Daily Estimates of Soil Ingestion in Children

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Soil ingestion estimates play an important role in risk assessment of contaminated sites, and estimates of soil ingestion in children are of special interest. Current estimates of soil ingestion are trace-element specific and vary widely among elements. Although expressed as daily estimates, the actual estimates have been constructed by averaging soil ingestion over a study period of several days. The wide variability has resulted in uncertainty as to which method of estimation of soil ingestion is best. We developed a methodology for calculating a single estimate of soil ingestion for each subject for each day. Because the daily soil ingestion estimate represents the median estimate of eligible daily trace-element-specific soil ingestion estimates for each child, this median estimate is not trace-element specific. Summary estimates for individuals and weeks are calculated using these daily estimates. Using this methodology, the median daily soil ingestion estimate for 64 children participating in the 1989 Amherst soil ingestion study is 13 mg/day or less for 50% of the children and 138 mg/day or less for 95% of the children. Mean soil ingestion estimates (for up to an 8-day period) were 45 mg/day or less for 50% of the children, whereas 95% of the children reported a mean soil ingestion of 208 mg/day or less. Daily soil ingestion estimates were used subsequently to estimate the mean and variance in soil ingestion for each child and to extrapolate a soil ingestion distribution over a year, assuming that soil ingestion followed a lognormal distribution. *Key words*: mass balance, risk assessment, soil ingestion, trace elements. *Environ Health Perspect* 103: 276–285 (1995)

The choice of trace elements in soil ingestion studies (*1–3*) has often been discussed, but nearly all published studies to date have reported estimates based on all trace elements ("tracers") evaluated in the study design (*4–7*). One exception is a study by Van Wijnen et al. (*8*), in which a single tracer was selected based on defining a concept of a limiting detectable value. Although this approach has some merit, the resulting estimates are likely to be negatively biased. Other authors have discussed advantages and differences among the tracers presented (*1*). These reports do not show any one tracer emerging as the criterion on which soil ingestion estimates should be based. The lack of a clear consensus on tracer choice has led to uncertainty in soil ingestion estimates.

We have offered some biologically and statistically based guidance on tracer selection (*1,9*). Basically, we proposed and developed a detection-limit model as a means of estimating the relative precision of soil ingestion estimates from a given tracer. This simple linear model relates the log mean-square error, in percent recovery, to the log of the food/soil ratio. The model is intuitive; larger values of food ingestion of an element, relative to the corresponding amount of the element ingested from a given quantity of soil, lead to more highly variable soil ingestion estimates. Although there are some limitations to this model (*10,11*), the approach presents a way to select trace elements. Using this model, we have concluded that zirconium, aluminum, and titanium are elements with optimal properties for estimating soil ingestion (*6*).

Study designs for most soil ingestion studies reported in the literature have included collection of fecal output and food intake daily from subjects from 3 to 7 days. Soil-ingestion estimates have been based on a mass balance equation, subtracting (when available) tracer amounts in food intake from amounts in comparable fecal output and then dividing by the tracer concentration in soil. Although food intake and fecal output have been collected daily in most studies, only the Amherst study (*6*) assessed the amount of the trace element in food and fecal samples on a daily basis. Other studies pooled the food samples and the fecal samples before chemical analysis (*7*). The daily assessment of tracers in food and feces has made it possible to estimate daily variability in tracer ingestion (*12*), but it has not been used before to form soil ingestion estimates.

In this paper, we present a methodology to construct daily soil ingestion estimates using daily food and fecal trace-element concentrations. The methodology directly links the physical passage of food and fecal samples and thus has the potential for eliminating variability in soil ingestion estimates based on transit time differences and irregular fecal samples. Using this methodology, we reanalyzed the data on soil ingestion for the Amherst study. The result of these analyses is a single soil ingestion distribution that effectively eliminates the issue of tracer selection in determining a soil ingestion estimate. Using these estimates and making the assumption that soil ingestion is log-normally distributed for a given subject over days of the year, we estimated soil ingestion for children from the Amherst study over an entire year.

Materials and Methods

Amherst Study and Construction of Average Daily Soil Ingestion

Our methods are specific for soil ingestion studies with daily determinations of trace elements in food intake and fecal output (*6*). In brief, a total of 64 children between the ages of 1 and 4 years were enrolled for 2 weeks. Each week, duplicate food samples were collected for 3 consecutive days, and fecal samples were collected for 4 consecutive days for each subject. The total amount of each of eight trace elements (aluminum, barium, manganese, silicon, titanium, vanadium, yttrium, and zirconium) was recorded for food and fecal samples collected. Day 1 for the food sample corresponded to the 24-hr period from midnight on Sunday to midnight on Monday of a study week. Day 1 of the fecal sample corresponded to the 24-hr period from noon on Monday to noon on Tuesday. Soil and dust concentrations of the trace elements were also reported. Results provided soil ingestion estimates based on trace-element concentrations in soil, trace-element concentrations in dust, and weighted averages of these concentrations (*6*). In this paper, we assume that all soil ingestion occurred from outdoor soil, and hence we base all soil ingestion estimates on trace element concentrations in soil. We made this assumption for simplicity and because of uncertainty regarding the relative portion of ingested soil from outdoor soil versus indoor dust (*13,14*).

To construct soil ingestion estimates for a trace element, we subtracted the amount of the trace element in food from that in fecal output and divided by the concentration of the element in soil. We express the amount of a trace element in food and in fecal output as a "soil equivalent," which is defined as the amount of the element in food (or fecal output) divided by the concentration of the element in soil. Using these definitions, subtracting the average food soil equivalent from the average fecal soil equivalent results in an estimate of soil ingestion for a trace element.

Previous element-specific estimates of soil ingestion have been estimated by subtracting an average daily food soil equivalent from an average daily soil equivalent fecal output (*6,7*). When we constructed these estimates for the Amherst study, averages were formed by dividing fecal output by 4 and dividing food intake by 3,

regardless of the amount of fecal output or food intake reported on a given day. Among the 128 subject-weeks reported, 4 days of fecal output were reported for only 50 subject-weeks, 40 subject-weeks reported fecal output on 3 days, 33 subject-weeks reported fecal output on 2 days, and 5 subject-weeks had fecal output reported only on 1 day during the 4-day fecal collection protocol period. The lack of fecal output on a given day was attributed to day-to-day variability and not to missed fecal samples (6). For this reason, the estimate of average fecal output was calculated by dividing all subject-weeks by 4.

This method of determining average daily estimates for food and fecal samples for a given week has some obvious shortcomings. For example, for 2 subject-weeks with a single day of fecal output, the fecal output occurred on the first day in the week. The food ingestion estimates on days 2 and 3 for these subject-weeks are of questionable relevance to the estimate of soil ingestion for the subject-week because they occurred after the fecal output. For 3 subject-weeks, fecal output samples were collected for the first 2 days in the study, but not for the last 2 days. Should the day-3 food intake (most of which occurred after the fecal output) be included in estimating the average daily food intake? Issues such as these motivate a more detailed evaluation of the method of calculating soil ingestion.

Construction of Daily Soil Ingestion Estimates for an Element

We propose a methodology to estimate soil ingestion on a daily basis that is directly connected to the passage of food to feces. The definition of daily soil ingestion depends on the assumed lag time between food intake and fecal output. This lag time varies from subject to subject, and it may vary with age or other subject characteristics. The lag time may also vary within a subject from week to week, or even from day to day. To illustrate the effect of lag time on soil ingestion estimation, let us define x_{ij} = as the soil equivalent ingestion from food for element j for the 24 hr period from midnight to midnight on day i, and y_{ij} as the soil equivalent ingestion from fecal samples for element j for the 24-hr period from noon on day i to noon on day $i + 1$, where $i = 1, \ldots , 4$ represents the 4 days of a subject-week, and $j = 1, \ldots , 8$ represents the eight trace elements included in the Amherst study. For example, assuming a lag time of 12 hr, the soil ingestion estimate for day i for element j is given by

$$y_{ij} - x_{ij} = \text{soil estimate (12-hr lag)}.$$

Assuming a 28-hr lag time, a soil ingestion

estimate for day i is given by

$$d_{ij} = y_{ij} - 1/3[x_{ij} + 2x_{(i-1)j}]$$
$$= \text{soil estimate (28-hr lag)}.$$

This estimate is equal to the soil equivalent of element j in the feces on day i, minus the soil equivalent of element j in food, where the food estimate is based on a 1:2 weighting of food in the two consecutive 24-hr periods ending at noon on day j.

If the lag times are known (or estimated) for each subject, estimates of daily soil ingestion estimates can be directly determined. If lag times are not available, daily soil ingestion can be estimated by making some assumptions about the lag times. We assume that for each subject, the lag time is constant for all days in a given week, and we assume that the lag period is uniform for all subjects and weeks, and equal to a 28-hr lag. These assumptions are consistent with the observation of a 1- to 4-day passage time for food among children (6). While the methodology has limitations (which we discuss later), it comes much closer to a biological representation of soil ingestion.

Two modifications are necessary to use this daily soil ingestion method. The first modification addresses the need to estimate food intake for days when duplicate food samples were not collected. The second modification addresses the way soil ingestion is estimated on days when no fecal output occurred.

Estimation of food intake. Fecal samples were collected in the Amherst soil ingestion study for 4 days (from noon Monday through noon Friday). Assuming a 28-hr food transit time, food samples would be required on Sunday through Thursday to estimate soil ingestion directly for each of the fecal output collection days. Unfortunately, the study protocol for food ingestion specified duplicate food samples only on Monday through Wednesday. Modification of the daily soil ingestion formula is necessary for days 1 and 4 because food ingestion was not reported for Sunday or Thursday. We use the mean soil equivalent food ingestion for a given subject-week as an estimate of food ingestion on Sunday and on Thursday. This results in soil ingestion estimates for Monday given by

$$d_{1j} = y_{1j} - 1/3[x_{1j} + 2 \bar{x}] = \text{soil estimate},$$

based on a 28-hr lag, and soil ingestion estimates for Thursday given by

$$d_{4j} = y_{4j} - 1/3[\bar{x} + 2x_{3j}] = \text{soil estimate}$$

based on a 28-hr lag.

Estimation of fecal output. A second modification is necessary for days when no fecal output was reported. True soil ingestion on a given day cannot be less than zero, and, ideally, the estimates of daily soil ingestion will be greater than or equal to zero for all days. Although actual soil ingestion cannot be negative, estimates of daily soil ingestion can be negative when the food intake exceeds the fecal output for a designated day. This can occur when the subject does not follow the assumed 28-hr time lag linking food tracer intake to fecal tracer output (e.g., if the fecal tracer output for the day corresponds to a different time window from the 24-hr period 28 hr before the start of the fecal day). Such a difference can result in a negative or positive bias in the soil ingestion estimate.

A single estimate of tracer ingestion of food was made each day, although meals for each day were not identical. This is a limitation of the soil ingestion study protocol, which sometimes results in negative or positive daily soil ingestion. If the portion of a day attributed to a fecal sample period has trace element ingestion from food lower than the daily tracer average for food, the soil ingestion estimate for the tracer will be negatively biased. A positive bias will result if the portion of the average tracer amount in food attributed to a fecal sample period is less than the actual tracer amount in food for the corresponding period.

Generally, it is inappropriate to reset negative soil ingestion estimates to zero when they occur, even though such negative soil ingestion is not physically possible. If we assume a subject consumes no soil, small positive and negative estimates of soil ingestion are likely to occur due to the imperfect linking of food tracer input and fecal tracer output samples resulting from the assumed 28-hr transit time period. On average, these estimates of soil ingestion will balance to zero. If negative soil ingestion estimates are reset to zero, a bias will be introduced in the final soil ingestion estimate because only the positive estimates will be counted. For this reason, negative soil ingestion estimates are not generally excluded nor reset to zero for a subject on a given day.

There is one situation, however, in which it is appropriate to alter negative soil ingestion estimates. Negative estimates of soil ingestion are clearly in error when they occur as a result of no fecal output on a given day. On such a day, the soil ingestion for that day cannot be assessed and is described as missing. For example, when no fecal output is observed for day(s) at the end of a subject-week, soil ingestion cannot be estimated for those day(s). The expected food soil equivalent for days with

no fecal output is called output error (15). Soil ingestion estimates for the subject-week are available only for the earlier week days with fecal output.

A different situation occurs when no fecal output is reported for a given day (1, 2, or 3) in the week, but fecal output is present for subsequent days in the week. In such a case, the mass balance equation has a gap (due to days with no fecal output), and the food intake for the corresponding period is unaccounted for. We make an adjustment to account for the food intake by adding the food intake for the no fecal output sample period to the food intake for the next occurring fecal sample. This adjustment corresponds to an assumption that, if no fecal output occurs on a given day, all food based tracers that would have been excreted for that day are contained in the next observed fecal sample period. To maintain the interpretation of the soil ingestion estimate as a daily estimate, we divide the estimated soil ingestion by 2 (if no fecal sample is reported for one day) or by 3 or 4, if either 2 or 3 consecutive days occur with no fecal samples reported, respectively.

Specifically, the adjustment implies that the day 4 soil ingestion estimate is calculated as:

No day 3 fecal sample:

$$d_{4j} = 1/2\{y_{4j} - 1/3[\,[\bar{x} + 2x_{2j}] - x_{3j}\}$$
where $d_{4j} = d_{3j}$;

No day 2 and 3 fecal samples

$$d_{4j} = 1/3\{y_{4j} - 1/3[\bar{x} + 2x_{1j}] - x_{3j} - x_{2j}\}$$
where $d_{4j} = d_{3j} = d_{2j}$;

No day 1, 2, and 3 fecal samples

$$d_{4j} = 1/4\{y_{4j} - \bar{x} - x_{3j} - x_{2j} - x_{1j}\}$$
where $d_{4j} = d_{3j} = d_{2j} = d_{1j}$.

A similar estimate may be determined when fecal samples for day 3 are present, but no fecal samples are reported for day 2 (or days 1 and 2):

No day 2 fecal sample

$$d_{3j} = 1/2\{y_{3j} - 1/3[x_{3j} + 2x_{1j}] - x_{2j}\}$$
where $d_{3j} = d_{2j}$;

No days 1 and 2 fecal samples

$$d_{3j} = 1/3\{y_{3j} - 1/3[x_{3j} + 2\bar{x}] - x_{2j} - x_{1j}\}$$
where $d_{3j} = d_{2j} = d_{1j}$.

Finally, if fecal samples for day 2 are present, but no fecal sample is reported for day 1, we estimate soil ingestion on day 2 as:

No day 1 fecal sample

$$d_{2j} = 1/2\{y_{2j} - 1/3[x_{2j} + 2\bar{x}] - x_{1j}\}$$
where $d_{2j} = d_{1j}$.

Best Estimate of Soil Ingestion for a Day for a Subject

Soil ingestion estimates can be calculated on a daily basis as outlined above. In the Amherst study, a maximum of eight such estimates can be determined for a given day, with one estimate per trace element. These estimates relate to a common time period for soil ingestion. If the 28-hr constant transit-time assumption is valid, this common time period is 24 hr (or 48, 72, or 96 hr if 1, 2, or 3 previous days had no reported fecal output). Since the trace-element estimates for a day estimate a common soil ingestion, we can compare the estimates and devise a criteria to select a best estimate. Our first step in establishing the best estimate is to estimate the median soil ingestion from among the eight element-specific estimates for the day. Next, we identify upper and lower bounds for a range of estimates based on criteria formed using assumptions on the relative standard deviation (RSD). We consider daily soil ingestion estimates for trace elements falling outside this boundary to be unreliable, and exclude them in subsequent calculations. Finally, we define the median of remaining trace element estimates as the best estimate of soil ingestion for the day for a subject.

Implementation of the methodology for determining the best daily soil ingestion estimate required us to specify criteria for identifying outlier estimates of soil ingestion. The median estimate of soil ingestion on day i for a subject-week is defined as d_i. We set the criteria for identifying unusual estimates of soil ingestion based on the difference, $\delta_{ij} = |d_{ij} - d_i|$. If δ_{ij} is larger than a given value, Δ_i, we consider the soil ingestion estimate based on element j unreliable, and hence exclude the trace-element estimate from the set of possible estimates of soil ingestion on that day.

The key in identifying unreliable soil ingestion estimates is determining Δ_i. We allow Δ_i to vary with the median estimate of soil ingestion in a manner determined by changes in the RSD, where we assume that

$$RSD_i = \Delta_i / \delta_i .$$

The values of Δ_i were determined so as to reflect our judgment of the limits of soil ingestion estimation. Basically, we considered the RSD to be at a minimum of 100% when the median soil ingestion estimate was less than 50 mg/day. With increasing soil ingestion, we assumed that the RSD would improve, becoming close to 50% for soil ingestion estimates of 500 mg/day. Finally, for very large soil ingestion estimates of 10 g/day, we assumed that the RSD was about 20%. With these

assumptions, a simple function was derived to approximate these assumptions. The resulting function is given by $\ln(RSD_i) = 1.5 - 0.35 [\ln(d_i)]$. Since low median estimates of soil ingestion result in artificially low values of Δ_i, we limited the minimum value of Δ_i to be 50 mg/day. As a result, we set

$$\Delta_i = \max (50, \ d_i e^{[1.5 - 0.35 \ln(d_i)]}).$$

Values of Δ_i for various estimates of median soil ingestion are given in Table 1. The difference between the ultimate median soil ingestion estimates for the subject-week-day, and soil ingestion estimates for elements judged to be outliers is termed source error (15), because it is thought to represent some other source of trace-element ingestion apart from food or soil.

Soil Ingestion Estimates for a Subject

Using the described methodology, daily soil ingestion estimates were calculated for each subject for up to 4 days for 2 weeks. The number of elements included in the final evaluation of the median soil ingestion varied for different subjects, weeks, and days. The number of days with soil ingestion estimates also varied for a subject. We consider the detailed construction of these estimates because these details relate to the validity and interpretation of the soil ingestion estimate. We describe several ways in which soil ingestion estimates can be calculated, to relate the resulting estimates to the previous literature, to make clear the limitations in previ-

Table 1. Criteria for identifying outlier estimates of soil ingestion[a]

d_i(mg/day)	Δ_i
20	50
50	57
100	89
150	116
200	140
250	162
300	183
350	202
400	220
450	238
500	255
1000	399
2000	627
3000	816
4000	984
5000	1137
6000	1280
7000	1415
8000	1543
9000	1666
10000	1784

[a] d_i = median (soil ingestion for day i) and Δ_i is a criteria such that if $|d_{ij} - d_i| > \Delta_i$, the estimate for element j is identified as an outlier estimate of soil ingestion.

ous soil ingestion estimates and to identify limitation in estimates presented in this paper.

In our model for soil ingestion, we represent soil ingestion, y, for subject s on day i with the measure of element j as

$$y_{sij} = \mu_{si} + e_{sij}$$

where μ_{si} is the "true" soil ingestion for subject s on day i, and e_{sij} is technical error, including error due to transit time and laboratory and preparation errors. We will assume that the days i correspond to days in a year. In this manner, a set of values μ_{si} for $i = 1, \ldots, 365$ will define the soil ingestion for subject s in the year. We define the subject's birthdate as the first day in the year for that subject. This definition is arbitrary, but it allows a simple description of age-specific soil ingestion. With these assumptions, subject s will have a different soil ingestion for each year of age.

For a given subject of a given age, we may summarize the set of daily soil ingestion in different ways. One parameter of interest is the total amount of soil consumed by the subject over the year (we use "year" to represent the set of days of a given age for the subject). Total soil ingestion is given by

$$\sum_{i=1}^{365} \mu_{si} .$$

We may also consider the average daily soil ingestion for the subject, defined as

$$\sum_{i=1}^{365} \frac{\mu_{si}}{365} ,$$

or the median daily soil ingestion estimate, given by

median $[\mu_{si}$ for all $i = 1, \ldots, 365]$.

The last two parameters are likely to reflect different characteristics of soil ingestion for the subject. We used estimates of these parameters to summarize results for the Amherst soil ingestion study.

Example of soil ingestion estimate construction for a subject. For each of the 64 subjects in the Amherst soil ingestion study, soil ingestion estimates can be classified in an 8 × 8 array of elements and study days. An example of such an array is presented in Table 2 for subject 849 and illustrates the methodology for soil ingestion estimation. For subject 849, no fecal samples were reported on days 1 or 4 during the first week or on day 4 during the second week. In the first week, fecal output on day 2 was used to estimate an average

soil ingestion over days 1 and 2. Estimates of this average daily soil ingestion were within the RSD boundaries for only two elements (silicon and zirconium), with a median soil ingestion estimate of 0.5 mg reflecting 2 days of soil ingestion, or 0.25 mg/day. No soil ingestion estimate could be made for day 4 in either week because of missing fecal samples. Soil ingestion estimates for two elements met the RSD criteria for day 3 in the first week, and, in the second week, four elements met the criteria for days 1 and 2 and five elements met the criteria for day 3. The median daily soil ingestion estimate ranged from 0.25 mg/day to 260 mg/day over the 6 days reported for subject 849.

Using the results shown in Table 2, we determined the median and mean daily soil ingestion estimate for subject 849; for the 6-day period for this subject the median soil ingestion estimate is 50.5 mg/day, with a mean of given 71.3 mg/day. Soil ingestion estimates for an element can also be constructed using the results in Table 2. Such estimates are inferior to estimates based on the median soil ingestion for the day because element-specific estimates are based on fewer days and are more likely to reflect positive or negative error. Element-specific estimates are similar, however, to previous estimates reported in the literature. When the previous day's fecal output is zero, the average daily estimate for an element is replaced by the estimate for the element when a fecal sample was reported (Table 2). The median element-specific estimates ranged from 10 to 82 mg/day, and the mean ranged from 24 to 148 mg/day. Note that all of the element-specific estimates of soil ingestion refer to a different set of subject days. If these days are considered a simple random sample of days for the subject, then the element-specific estimates can be considered estimates of a common parameter corresponding to median (average) daily soil ingestion for the subject.

Soil ingestion estimates can also be determined for each element for the week. When forming the weekly estimates, if all 4 days of soil ingestion are available, the weekly estimate of average daily soil ingestion is given by the simple average of the daily estimates. Note that even with complete data, this average may differ from the median daily soil ingestion estimate because soil ingestion estimates are skewed. Although weekly soil ingestion estimates have been presented in previous publications (6,7), there appears to be no conceptual advantage to forming such estimates over the median and mean estimates.

Results

Estimation of Daily Soil Ingestion

The results for the 64 children included in the Amherst study (6) are based on daily estimates of soil ingestion. Although all 64 subjects participated in the study for a total of 8 days, it was not possible to estimate soil ingestion for all 8 days for each subject due to missing fecal samples. Table 3 shows the distribution of the number of days of soil ingestion estimates for the subjects overall and by tracer elements after eliminating estimates based on the outlier criteria given in Table 1. Daily soil ingestion estimates (based on the median of the daily soil ingestion estimates) were present for at least 4 days for all subjects and were present for 6 or more days for 94% of the subjects. For aluminum, soil ingestion estimates were available for all subjects on at least 2 days. No other trace element satisfied inclusion criteria for soil ingestion estimation for all subjects for at least 1 day. However, silicon, zirconium, and yttrium satisfied the inclusion criteria on at least 1 day for 63, 62, and 61 subjects, respectively. For barium and manganese, a large proportion of subjects (48% and 70%, respectively) had no days with "valid" soil ingestion estimates (that did not exceed the outlier criteria).

Table 2. Soil ingestion estimates (mg/day) for subject 849 in the Amherst study

Element (j)	Week 1				Week 2				Element-specific	
	Day 1[a]	Day 2	Day 3	Day 4	Day 1	Day 2	Day 3	Day 4	Median	Mean
Al (1)					17	54	65		54	45
Ba (2)										
Mn (3)										
Si (4)		2			77	127	98		77	61
Ti (5)			414		68	55	53		54	148
V (6)						78			78	78
Y (7)			105				59		82	82
Zr (8)		-1			22		77		10	24
N (elements)	0	2	2	0	4	4	5	0		
Daily soil ingestion[c]	0.25	0.25	260	—[b]	46	55	66	—[b]		

[a]Day = i.
[b]No daily soil ingestion estimate because there was no subsequent fecal sample.
[c]Given by the median of the soil ingestion estimates for a given day.

Table 3. Distribution of number of daily soil ingestion estimates per subject overall and by element[a]

No. of estimates	Overall	Al	Ba	Mn	Si	Ti	V	Y	Zr
0	0	0	31	45	1	8	12	3	2
1	0	0	8	8	2	4	9	1	0
2	0	6	13	6	3	9	7	2	3
3	0	7	6	5	6	6	8	12	10
4	1	8	4	0	7	12	8	13	8
5	3	13	2	0	18	11	8	7	13
6	21	17	0	0	12	9	6	12	17
7	18	7	0	0	8	1	4	11	7
8	21	6	0	0	7	4	2	3	4

[a]Table entries are the number of subjects with the given number of days of soil ingestion. For example, under the "overall" column, the value of 21 at the bottom of the column means that 21 subjects had 8 days on which soil ingestion estimates were provided.

Figure 1 (using a $\log_{10}$ scale for soil ingestion) and Figure 2 (using an arithmetic scale for soil ingestion) summarize the cumulative distribution of daily soil ingestion estimates, including estimates of the median daily soil ingestion per child (M), and the average daily soil ingestion per child (A). The average soil ingestion estimate for one subject (who had an average estimate of 7.7 g/day of soil ingestion) is not included in the figures because inclusion alters the scale. For half the children, the median daily soil ingestion estimate was less than 12 mg/day, while the mean soil ingestion estimate was less than 45 mg/day. Similarly, for 95% of the children, the median daily soil ingestion estimate was less than 138 mg/day, while the mean daily soil ingestion estimate was less than 208 mg/day. Tables 4 and 5 contain summaries of certain percentile estimates based on the median and mean daily soil ingestion estimates. Estimates based on barium and manganese are unreliable in part due to large soil equivalent ingestion
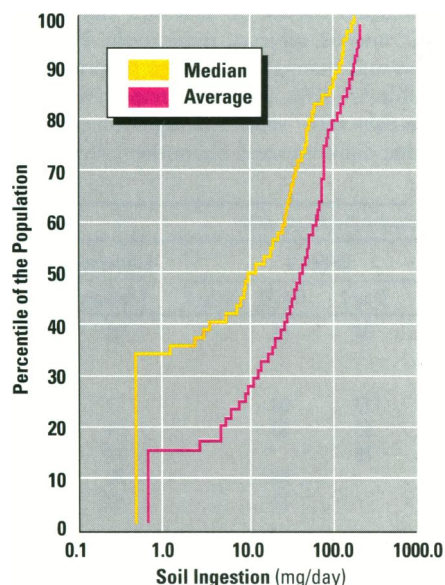
from food. Trace element-specific estimates based on days where the trace element estimate satisfied the daily inclusion criteria are provided in Tables 4 and 5 for comparison with previous published estimates, but are not recommended for use.

As previously mentioned, estimates of soil ingestion for all 8 days were not made for all subjects (see Table 3). Furthermore, on a given day when a soil ingestion estimate was made for a subject, the estimate was not necessarily based on all eight trace elements because element estimates that exceeded the outlier criteria were excluded. The number of days with soil ingestion estimates depended on the pattern of daily fecal samples and ranged from 4 to 8. The
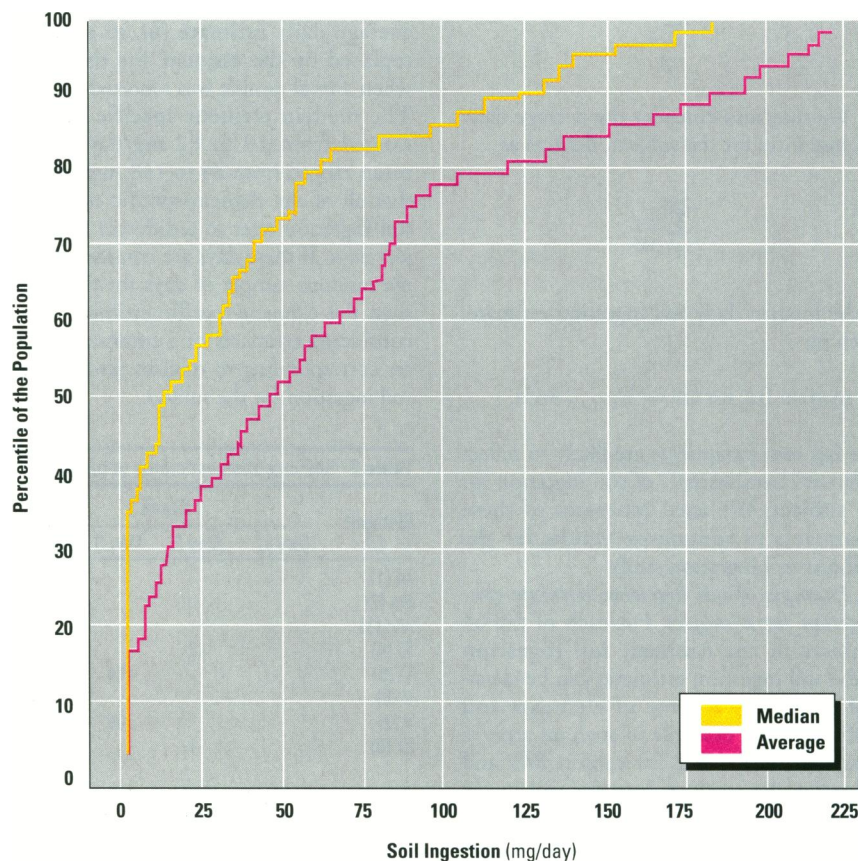
number of elements used in forming a daily estimate depended on how many estimates fell within the RSD window (Δ); this number also varied from one to eight. When a large number of element-specific estimates satisfied inclusionary criteria for each day and calculation of estimates for a large number of days was possible, we expected the soil ingestion estimate to be more reliable. In contrast, less reliable estimates of soil ingestion were expected for a subject when few elements and few days contributed to the estimate.

We used this reasoning to evaluate the sensitivity of the soil ingestion estimates to the number of daily estimates for a subject and the number of element estimates for a day. First, we recalculated soil ingestion estimates only if a minimum number of element-specific estimates were within the RSD window. Next, we considered soil ingestion estimates only for subjects with a minimum number of days of soil ingestion estimates.

Table 6 summarizes soil ingestion estimates when daily estimates are calculated only for days with a minimum number of trace element estimates. The number of days and subjects represented in these estimates varies by row due to application of the criteria for identifying outlier ingestion estimates. Table 6 includes estimates of soil

Figure 1. Cumulative distribution of soil ingestion estimates (median and mean) for 64 children (using $\log_{10}$ scaling for soil ingestion; subject exhibiting pica not displayed).

Figure 2. Cumulative distribution of soil ingestion estimates (median and mean) for 64 children (using arithmetic scaling for soil ingestion (subject exhibiting pica not displayed).

ingestion when no element-specific estimates were excluded via the outlier criteria. Table 6 also summarizes characteristics of the subjects described by the medium and mean soil ingestion estimates. The minimum number of element-specific soil ingestion estimates included in a daily soil ingestion estimate varies from 1 to 8. All 64 subjects have at least 1 day with 3 or more eligible element-specific estimates. When the criteria for number of element estimates per day is larger than 4 and the outlier criteria are used, the number of subjects with soil ingestion estimates decreases. Due to missing fecal samples on days at the end of study weeks for some subjects, soil ingestion estimates were calculated for only 86% (439) of the possible subject-days in the study protocol (8 days × 64 subjects = 512 or 100%). The proportion of days with soil ingestion estimates decreases as the criteria for inclusion are made more stringent. The decrease is modest when criteria require three or more eligible element-specific estimates. When four or more element-specific estimates are required, the number of days with soil ingestion estimates decreases markedly. The average number of days in which a soil ingestion estimate can be made for a subject (Table 6) decreases when more than three element-specific estimates are required.

A summary of the distribution of median daily soil ingestion estimates and mean daily soil ingestion estimates for subjects, based on the different criteria for number of element-specific estimates per day, is also presented in Table 6. Values in this table are comparable only when the number of subjects is the same. As a result, comparison of 1, 2, 3, and 8 trace elements per day (based on 64 subjects) provides a measure of sensitivity of the soil ingestion distribution to the minimum number of trace element estimates required to estimate soil ingestion for a day. This comparison indicates generally that estimates of the median daily soil ingestion distribution is slightly less sensitive to the number of estimates than the mean soil ingestion distribution and that larger differences in estimates occur for higher percentiles of the distributions. When subject numbers are varied, interpretation of comparisons is complicated because different children contribute to the estimates.

Based on the results of Table 6, we chose to evaluate the effect of different numbers of daily soil ingestion estimates using a minimum of three eligible trace-element estimates per day to form a daily estimate. We selected 3 eligible element estimates because all 64 subjects were included, and because there was a relatively large number of days with soil ingestion estimates per subject. We varied the minimum number of daily estimates from 2 to 7 because all subjects had at least 2 days of soil ingestion estimates. The results are summarized in Table 7.

Evaluating the effect of increasing the minimum number of days to form a soil ingestion estimate for a child is complicated by the different numbers of subjects included, based on different criteria for the days. When a minimum of 2–4 days of soil ingestion estimates are required, more than 90% of the subjects have soil ingestion estimates, and the distribution of mean

Table 4. Distribution of median daily soil ingestion estimates per child (mg/day) for 64 children[a]

Type of estimate	Overall (64)[b]	Al (64)	Ba (33)	Mn (19)	Si (63)	Ti (56)	V (52)	Y (61)	Zr (62)
Mean	32	32	678	1039	31	217	101	15	19
25th Percentile	0	0	7	35	0	0	4	0	0
50th Percentile	13	13	52	121	12	15	24	6	11
75th Percentile	50	37	233	313	65	57	133	44	34
90th Percentile	126	76	473	355	99	147	372	91	90
95th Percentile	138	137	562	17,416	164	279	481	106	121
Maximum	185	411	19,068	17,416	387	10,086	845	226	167

[a]For each child, estimates of soil ingestion were formed on days 4–8 and the median of these estimates was then evaluated for each child. The values in the column "overall" correspond to percentiles of the distribution of these medians over the 64 children. When specific trace elements were not excluded via the relative standard deviation criteria, estimates of soil ingestion based on the specific trace element were formed for 1–8 days for each subject. The median soil ingestion estimate was again evaluated. The distribution of these medians for specific trace elements is shown.
[b]n in parentheses.

Table 5. Distribution of average (mean) daily soil ingestion estimates (mg/day) per child for 64 children[a]

Type of estimate	Overall (64)[b]	Al (64)	Ba (33)	Mn (19)	Si (63)	Ti (56)	V (52)	Y (61)	Zr (62)
Mean	179	122	655	1,053	139	271	112	165	23
25th Percentile	10	10	28	35	5	8	8	0	0
50th Percentile	45	29	65	121	32	31	47	15	15
75th Percentile	88	73	260	319	94	93	177	47	41
90th Percentile	186	131	470	478	206	154	340	105	87
95th Percentile	208	254	518	17,374	224	279	398	144	117
Maximum	7,703	4,692	17,991	17,374	4,975	12,055	845	8,976	208

[a]For each child, estimates of soil ingestion were formed on days 4–8 and the mean of these estimates was then evaluated for each child. The values in the column "overall" correspond to percentiles of the distribution of these means over the 64 children. When specific trace elements were not excluded via the relative standard deviation criteria, estimates of soil ingestion based on the specific trace element were formed for 1–8 days for each subject. The mean soil ingestion estimate was again evaluated. The distribution of these means for specific trace elements is shown.
[b]n in parentheses.

Table 6. Summary of distribution of daily soil ingestion estimates (mg/day) per child based on a minimum number of trace elements

Trace element estimates/ day	No. of subjects	% of days[a]	Average no. of days/ subject	Median (all children)[b]							Mean (all children)						
				Mean	25th	Median	75th	90th	95th	Max	Mean	25th	50th	75th	90th	95th	Max
1+	64	86	6.9	32	0	13	50	126	138	185	179	10	45	88	186	208	7,703
2+	64	85	6.8	36	0	13	50	130	161	357	182	10	45	88	186	210	7,703
3+	64	75	6.0	26	0	11	49	71	112	226	164	10	34	76	112	151	7,703
4+	63	58	4.7	175	0	9	46	73	146	9,649	243	3	24	54	93	148	13,306
5+	53	36	3.4	210	0	12	31	72	259	9,622	214	0	17	43	97	259	9,622
6+	28	14	2.6	717	3	18	34	134	226	19,261	721	4	19	43	134	226	19,260
7+	6	2	1.8	101	18	77	198	226	226	226	98	18	66	198	226	226	226
8[c]	64	86	6.9	38	0	14	71	150	187	324	157	9	48	116	166	210	6,422

[a]Total of 512 days.
[b]Percentiles indicated by 25th, 75th, 90th, and 95th.
[c]No elements excluded via outlier criteria.

Table 7. Summary of distribution of daily soil ingestion estimates (mg/day) per child based on a minimum of three trace element estimates per day, with varying minimum numbers of daily estimates

No. of days with soil ingestion estimates/ day	No. of subjects	% of days[a]	Average no. of days/ subject	Median (all children)[b]							Mean (all children)						
				Mean	25th	Median	75th	90th	95th	Max	Mean	25th	50th	75th	90th	95th	Max
2+	64	75	6.0	26	0	11	49	71	112	226	165	10	34	76	112	151	7703
3+	63	74	6.0	26	0	11	48	71	112	226	167	9	34	77	116	151	7703
4+	59	72	6.3	20	0	10	38	68	107	119	171	7	32	77	112	151	7703
5+	54	68	6.5	20	0	10	37	61	107	119	181	7	30	63	112	151	7703
6+	48	62	6.6	20	0	11	35	61	94	112	200	8	30	61	119	151	7703
7+	21	31	7.5	15	0	9	20	38	50	112	391	5	21	44	78	107	7703

[a]Total of 512 days.
[b]Percentiles indicated by 25th, 75th, 90th, and 95th.

and median estimates is relatively stable. This result suggests that the estimated soil ingestion distribution is not greatly affected by subjects with more limited soil ingestion data.

Annual Soil Ingestion Distribution Estimates Based on Daily Soil Ingestion Values

Individual daily soil ingestion estimates summarized in Tables 4 and 5 were used to develop a distribution of values for 365 days for each subject using an assumed log-normal distribution. Between 4 and 8 daily soil ingestion estimates were available on each of the 64 subjects. By assuming that soil ingestion is log-normally distributed for a given subject, we estimated the parameters for the log-normal distribution based on the subjects' daily estimates (replacing negative daily soil ingestion estimates by an estimate of 1 mg/day). Order statistics, corresponding to z-scores for percentiles in increments of 1/365, were then used in connection with the log-normal distribution and estimated parameters to form soil ingestion estimates for 365 days for each subject. Characteristics of these annual soil ingestion distributions are summarized in Tables 8 and 9. Of particular note is that the estimated median of the 64 subjects' daily soil ingestion as averaged over 365 days is 75 mg/day, while the upper 95% is 1751 mg/day. The findings also indicate that more than 10% of the subjects ingest on the average approximately 1.2 g/day. Table 9 indicates that 33% of the children will ingest >10 g of soil on 1–2 days per year, and 16% of children are expected to ingest >1 g of soil on 35–40 days per year.

Figure 3 (using a log_{10} scale for soil ingestion) and Figure 4 (using an arithmetic scale for soil ingestion) summarize the cumulative distribution of daily soil ingestion estimates for the 64 subjects, including estimates of the median daily soil ingestion per child (M) and the average daily soil ingestion per child (A). In contrast to Figures 1 and 2, the estimates plotted in Figures 3 and 4 are based on predicted soil ingestion over an entire year (365 days), assuming a log-normal soil ingestion distribution. The results clearly demonstrate a marked increase in the cumulative distribution for soil ingestion using subject averages. This result can be expected based on the skewness of the assumed log-normal soil ingestion distribution.

Discussion

Daily Soil Ingestion Ingestion Framework

The results presented here represent the first estimates of daily soil ingestion based on a mass–balance study that have appeared in the literature. Previous reports have estimated average daily soil ingestion

Table 8. Soil ingestion estimates on 64 subjects over 365 days based on fitting a log-normal distribution model to daily soil ingestion values

Range of median soil ingestion estimates of 64 subjects over 365 days	1–103 mg/day
Median soil ingestion of 64 subjects[a]	14 mg/day
Range of upper 95% soil-ingestion estimates of 63 subjects over 365 days[b]	1–5263 mg/day
Median of the upper 95% soil ingestion estimates of the 64 subjects over 365 days	252 mg/day
Range of estimated total number of grams of soil ingested per year by 63 subjects[b]	0.365–828.16 g
Range of average daily soil ingestion values for the 63 subjects over 365 days[b]	1–2268 mg/day
Median of the 64 subjects daily average soil ingestion[a]	75 mg/day
The upper 95% of the average daily soil ingestion for 64 subjects[a]	1751 mg/day
The upper 90% of the average daily soil ingestion for 64 subjects[a]	1190 mg/day

[a]With subject estimates equal to the average soil ingestion over 365 days.
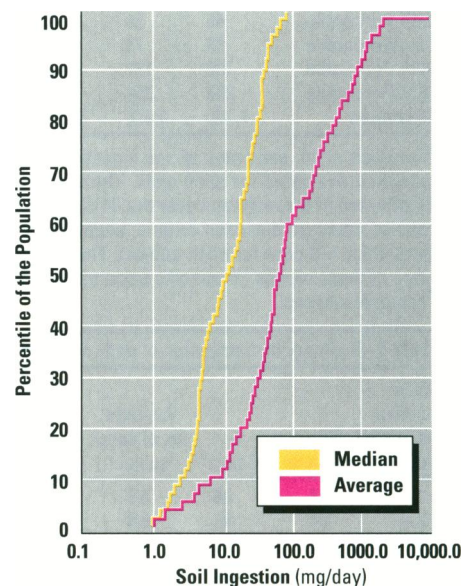[b]Subject exhibiting soil pica excluded.

where the average was taken over 3, 4, or 8 days for a given subject (6,7). Since soil ingestion estimates are likely to be positively skewed for a subject, the distribution of average daily soil ingestion estimates (with averages taken over a specified period of time) will be larger than the distribution of daily soil ingestion estimates reported here. This effect is observable in the difference between the soil ingestion distributions summarized in Tables 4 and 5.

Table 9. Estimated percent of children with soil ingestion exceeding daily rates for given time periods per year

Estimated no. of days/year with soil ingestion	Daily rate of soil ingestion				
	>200 mg	>500 mg	>1 g	>5 g	>10 g
1–2	86	72	63	42	33
7–10	72	53	41	20	9
35–40	42	31	16	1.6	1.6

Figure 3. Cumulative distribution of soil ingestion estimates (median and mean) for 64 children (using a log_{10} scaling for soil ingestion) based on soil ingestion estimated over 365 days based on a log-normal distribution (subject exhibiting pica not displayed).

Overall soil ingestion estimates presented in Tables 4 and 5 are not trace-element specific. Furthermore, these soil ingestion estimates are robust to sample loss and source errors affecting individual trace element estimates, and they account for positive/negative error (15). The daily soil ingestion methodology directly links physical passage of food and fecal samples and thus has the potential for eliminating variability in soil ingestion estimates based on transit time differences and irregular fecal samples. For these reasons, we consider the daily soil ingestion methodology to be superior to other strategies for understanding and quantifying soil ingestion.

Previous studies have considered element-specific soil ingestion estimates. A summary of the distribution of these estimates relative to the daily estimates presented in Table 5 is given in Table 10. In the absence of biases or positive/negative errors, the estimates based on the two methods should be the same. Differences in the estimates reflect differences due to

sources, as well as different numbers of subjects contributing to the estimates based on the daily method. It is noteworthy that daily estimates were possible for all subjects for aluminum and for all but one subject for silicon. The distribution of the daily estimates and the distribution of the original estimates are quite similar, indicating little error due to these possible sources. This suggests that if element-specific estimates were used, aluminum and silicon should be selected. This is not to say that the estimates based on either of these elements is preferable to the overall estimate based on all elements. Individual trace-element estimates are more subject to input, output, and source errors (15), all of which are addressed and potentially minimized using the daily methodology.

Although the daily soil ingestion estimates offer a substantial improvement over element-specific estimates, these estimates depend on several assumptions. Perhaps the most suspect assumption is that children's fecal samples can be grouped on a

daily basis and directly linked to a 24-hr food sample for the period 28 hr before the fecal sample collection. We assume that this lag period is constant for all subjects and for all days, but the impact of this assumption on soil ingestion estimates has not been evaluated. It is possible that use of a constant 28-hr lag time coupled with irregular daily fecal output may positively bias soil ingestion estimates on the first day of a soil ingestion study due to several days ingestion contributing to the first day fecal sample. Using available data and additional assumptions, such as a constant fecal mass/food freeze-dry rate ratio, it may be possible to estimate lag times for individual subject-weeks. Other strategies may be possible to tailor lag times to individual subject-days. Sensitivity analyses of the 28-hr lag assumption should also be conducted.

An additional assumption used in this paper is the outlier criteria used to eliminate unusual element-specific estimates for individual subject days. Such a criteria was used due to the strong likelihood that
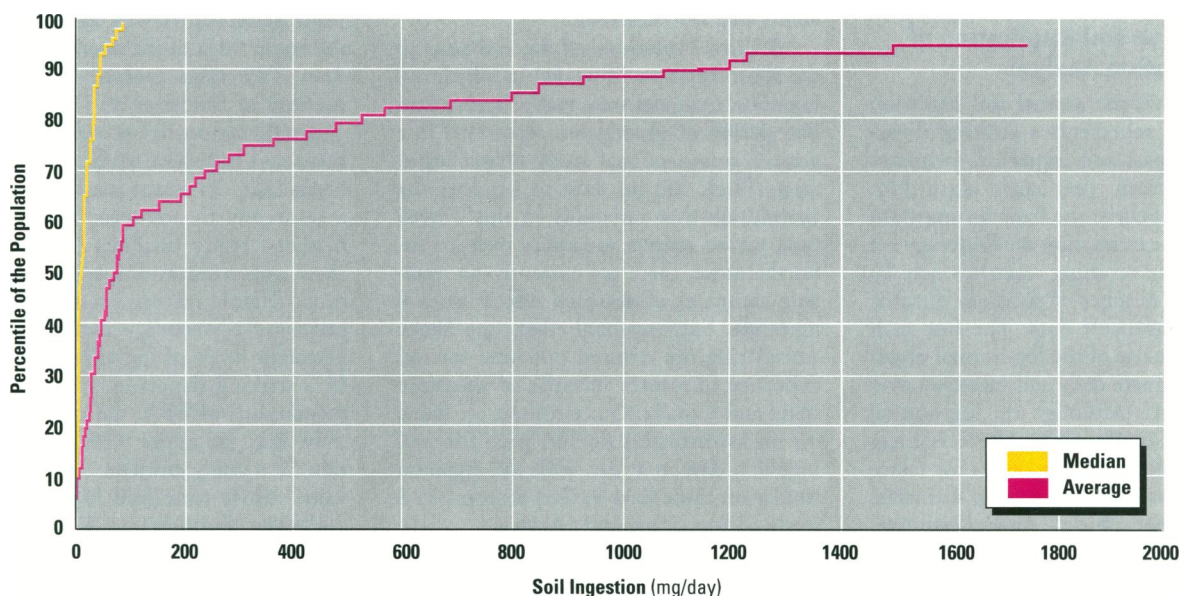

Figure 4. Cumulative distribution of soil ingestion estimates (median and mean) for 64 children (using a natural scale for soil ingestion) based on soil ingestion estimated over 365 days based on a log-normal distribution (subject exhibiting pica not displayed).

Table 10. Comparison of the distribution of the average (over all days with a soil ingestion estimate) daily soil ingestion estimates (mg/day) per child with the original element-specific estimates for 64 children

Type of estimate	Source[a]	Overall (64)	Al (64,64)[b]	Ba (33,64)	Mn (19,64)	Si (63,64)	Ti (56,64)	V (52,62)	Y (61,62)	Zr (62,62)
Mean	Daily	179	122	655	1,053	139	271	112	165	23
	Original		153	32	-294	154	218	459	85	21
50th percentile	Daily	45	29	65	121	32	31	47	15	15
	Original		29	-37	-261	40	55	96	9	16
90th percentile	Daily	186	131	470	478	206	154	340	105	87
	Original		138	228	595	219	702	1,366	91	67
95th percentile	Daily	208	254	518	17,374	224	279	398	144	117
	Original		223	283	788	276	1,432	1,903	106	110

[a]Daily soil ingestion estimates correspond to average estimates from Table 5. Original soil ingestion estimates correspond to estimates for 8 days based on original calculations [see Calabrese et al. (18.Table 13)].
[b]Numbers in parenthese indicate n_1 and n_2, respectively. Note that n_1 indicates the number of subjects contributing to the daily estimate.

ingestion of some tracers from sources other than food or soil occurred (16). This criteria helped to focus attention on discrepancies between individual trace-estimates of soil ingestion (Table 3). While helpful in this respect, the use of the outlier identification assumptions are not critical to the main soil ingestion results of the paper, as evident in comparing the overall results in Tables 4 and 5 with results in Table 6 (where no outliers were excluded).

A final limitation of the methodology and results is that, for each subject and day, a single estimate has been used for soil ingestion, even though multiple estimates were available from different trace elements. The multiple estimates allow both a point estimate and an estimate of the variance to be made on a subject-day basis. We have presented the point-estimate, but we did not assess the variance in these point estimates on a subject-day basis. Since accounting for this variance is likely to alter the estimated predicted distribution of soil ingestion, further analysis in this area is warranted.

Interpretation and Application of the Distribution Analysis

The analysis of the annual soil ingestion distributions represents a striking divergence from past soil ingestion estimates generated from the same data (6). Although we believe the basic assumptions underlying the estimation methodology are reasonable, the resulting estimates may be sensitive to a number of decisions dictated by limits in available data. The first such decision is the use of the log-normal distribution to estimate daily soil ingestion over a calendar year. Although the log-normal distribution is plausible, there is little empirical evidence to support its use (since only 4–8 days of soil ingestion estimates are available per subject). Annual estimates of total soil ingestion will be strongly affected by the tails of the soil ingestion distribution, which are uncertain. While log-normal modeling has been commonly used in exposure quantification practices (17), assumptions of alternative right-skewed distributions may produce large differences in total soil ingestion estimates. Data currently available are not adequate to resolve this issue.

Extrapolation of soil ingestion over a year is based on estimated parameters for the log-normal distribution. Assuming unbiased daily soil ingestion estimates, estimates of the mean and variance of the log-normal distribution have large variance because only four to eight measures are available for a subject. Variability in the variance estimates will generate large variability in annual soil ingestion estimates. This variability has not been assessed. A

further factor may serve to positively bias variance estimates for the log distribution. The methodology for estimating daily soil ingestion assumed a constant transit time of 28 hr. Variation in this transit time from day to day may result in inducing variability in daily soil ingestion estimates (falsely) and hence lead to higher variance estimates for the daily soil ingestion distributions. Such an error would act as a positive bias on annual soil ingestion estimates. At present, there is no information on the extent of transit-time error. However, if transit time error is large, we can expect fewer tracer-specific estimates of soil ingestion to satisfy the outlier criteria because tracers with more variable food intake result in highly variable soil ingestion estimates. The extent to which the soil ingestion distribution is stable when varying the number of required element estimates for a day is an indirect measure of potential bias due to transit-time error. The results of such an analysis (Table 7) suggest that the soil ingestion distribution is relatively stable and that a bias due to transit-time variability may not be severe.

A third limitation of the estimates is the short duration (2 weeks) in which soil ingestion estimates were made. Such a limited period of observation represents in a sense a cross-sectional study of soil ingestion. There are no data to support the assumption that variability in this 2-week time period reflects variability over a year. Furthermore, there are currently no available estimates of seasonal effects on soil ingestion. Such seasonal effects are likely, considering the reduced potential for soil exposure in winter months in northern states and Canada. Our estimates are based on an assumption of 365 possible soil ingestion days in a year, with soil ingestion similar on those days to that observed in 2 weeks in September and October.

Finally, the results are limited by the study protocol for the study (6,18). The children studied were a nonrandom sample residing in and adjacent to an academic community in western Massachusetts. Whether the soil ingestion behavior of these children is quantitatively relevant to population groups in other geographical regions (e.g., the inner city, rural areas) is unknown.

Despite these limitations, we consider our results to be extremely important for assessing exposure via soil ingestion. The results are based on a biologically refined model that fits physical soil ingestion kinetics. Variability due to the principal types of positive and negative error is likely reduced. Tracer-specific discrepancies are minimized. Daily soil ingestion estimates are calculated based on a physical mass-balance equation. Each of these aspects leads

to fundamentally improved individual and population-based soil ingestion estimates. The results provide a foundation for a more realistic appraisal of soil ingestion.

Our results suggest that soil pica in the general population needs to be reassessed. The current assessment indicates that soil pica is not merely truncated in a small subgroup, but that most children will periodically display this behavior to varying degrees of potential public health concern throughout the year. In addition, soil ingestion behavior may be an important factor in the characterization of acute and chronic effects.

An important question that needs to be addressed is how do the present analyses relate to current recommendations for soil ingestion exposure factors? Current practice has followed the guidance offered by the U.S. Environmental Protection Agency (EPA) and has been principally concerned with assessing public health risks from exposure to toxic substances at contaminated sites. EPA-recommended soil ingestion rates for children are 200 mg/day, a value generally viewed as approximating the upper 95% of the distribution for children. This value evolved from the initial findings of Binder et al. (4) and has been generally supported in subsequent childhood soil ingestion studies in the United States (6,7). In recent years, there has been a tendency to advocate the use of the Amherst study findings (19) in light of their improved attempts to identify the most reliable tracers based on estimated precision of recovery and soil ingestion detection levels of individual tracers (1,9). In a striking departure from the recommendations of EPA, the present findings show that the upper 95% of the distribution for a yearly estimate is 1750 mg/day, a value nearly nine-fold higher than EPA guidelines. Furthermore, it is possible that these estimates are approximately 5–10% low due to missing daily fecal samples as a result of incomplete subject compliance and a decision not to collect fecal material adhering to diaper wipes or toilet paper. Because soil ingestion is often a driving factor in the risk assessment process for contaminated sites, the implications of the current findings are likely to be substantial in terms of both estimated human health risks and in site-remediation costs.

Finally, it is important to recognize that the capacity to design, conduct, assess, and interpret soil ingestion studies has markedly improved since the first quantitative attempt (4). Since that time, methodological developments have given researchers the capacity to derive study-specific soil ingestion detection limits (1,9), differentiate soil from dust ingestion (13), identify, quantify, and correct for error (15), and apply these

approaches to the level of subject-day so that distributions of soil ingestion can be derived for each subject, as in the present paper. These developments now permit a more sophisticated approach to the estimation of soil ingestion estimates than was available several years ago. In addition, the application of such methods will result in the derivation of more defensible estimates of soil ingestion.

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