VARIATION IN TRANSFER FACTORS FOR STOCHASTIC MODELS: SOIL-TO-PLANT TRANSFER

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Abstract—**The scientific literature is a good source of information about soil-to-plant transfer, usually documented as mean concentration ratios. It does not provide, however, good information about variation in concentration ratio values, because most studies are done with limited numbers of crops and soils. Fortunately, a suitably large and diverse database is available from the International Union of Radioecologists that can be used to investigate variation. It has a total of over 7,000 concentration ratio values for over 22 elements. This paper reports on analysis of the geometric standard deviations obtained from the database. We assert that geometric standard deviation values are comparable and transferable among elements, so that geometric standard deviations calculated from large databases can be applied to other elements when there is a paucity of data. For a fully generic situation, where neither crops nor soil types are known, the geometric standard deviations is typically about 6. This implies a 1,300-fold range to encompass 95% of the data. For concentration ratios obtained for crops of interest on a specific site, the geometric standard deviation values are much lower, about 1.5. This still implies a fivefold range to encompass 95% of the data. Other values of geometric standard deviations for intermediate levels of information are provided. These estimates of geometric standard deviation are important to and appropriate for stochastic simulations of the impacts of soil contamination on crops and subsequent food chains. They are also important to the inclusivity of concentration ratios value distributions for unusual but perhaps important crops and settings.**
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Key words: soil; transfer coefficient; modeling, environmental; food chain

INTRODUCTION

ENVIRONMENTAL IMPACT assessments frequently have to estimate conditions far into the future or for settings that are otherwise not well known. One approach is to use stochastic modeling techniques. In these, the range of possible conditions is estimated with simulations that incorporate the expected variation in the underlying parameters. To do this requires information not only on the most likely or median value for each parameter, but also on the variation, dependence, and limit values for each parameter. Unfortunately, although there has been a traditional, strong emphasis on determining and documenting the median values and dependencies, there has been little consistent emphasis on documenting variation and limit values. This is especially a deficiency when there is a need for upper-fractile parameter values to use in sensitivity and worst-case analyses.

A major difficulty in reporting useful measures of parameter variation is to obtain enough data from independent sources so that the measured variation is a reasonable estimate of the population variation. Apart from geographically dispersed surveys such as Sheppard and Evenden (1990), most studies of soil-to-plant transfer of contaminants provide data for only a few soil types or a few crop types. Fortunately, the International Union of Radioecologists (IUR) has, over about ten years, compiled a large database of soil-to-plant transfer factors. These values are referred to here as concentration ratios (CRs). For cesium, strontium, cobalt, plutonium and neptunium there are 2,035, 920, 782, 679 and 465 records, and there are further records for americium, cerium, curium, iodine, lanthanum, manganese, nickel, lead, radium, ruthenium, antimony, technetium, thorium, uranium and zinc. The database was scrupulously maintained, under the direction of M. Frissel, to avoid inappropriate data. Each data entry is accompanied by information on crop, soil type, and experimental details. It is an excellent resource for examining variation of the CRs, and that is the objective of this paper. A specific application also discussed is the variation in CR values used in the assessment of the Canadian nuclear fuel waste management (NFWM) concept (Zach and Sheppard 1991; Zach et al. 1994).

Definition and statistical characteristics of CR

The soil-to-plant CR is defined by the IUR as the concentration of an element in the plant, on a dry weight basis, divided by that in the soil, also on a dry weight basis. Typically, the concentration in the plant part consumed by humans or livestock is reported. In this IUR database, the soil concentration was for the top 20 cm, except for grass where it was the top 10 cm. As a model of soil-to-plant transfer, CRs are entirely empirical and hence self-validating. Values of CR are considered to be element-specific, although quite often they are measured for radioactive isotopes of the elements. The total con-

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centration of the isotope measured in the soil is reported, regardless of bioavailability. For most of the data in the database, the radioisotope measured had been recently applied and, therefore, was relatively bioavailable compared to stable isotopes of the same element occluded in soil particles. Information on the source of the radioisotope in the soil was included in the IUR database.

Ratios such as CRs have a tendency, formalized by the Central Limit Theorem, to be lognormally distributed. This is also found to be true empirically, and is generally true for the CR values in the IUR database. As a result, geometric means (GMs) and geometric standard deviations (GSDs) are appropriate to summarize the data. The GSDs have fortunate attributes: 1) they are unitless and do not change value even if the CR is expressed on a plant wet weight basis, as done in some assessments, and 2) they are directly comparable among elements even though bioavailability varies markedly among the elements. These attributes are especially useful because it is possible to obtain good estimates of GSD from large databases and then apply them to elements or settings where there is a paucity of data.

In stochastic models, input parameters may be represented by probability density functions (PDFs), where a probability is assigned to each possible value of the parameter. For a lognormally distributed parameter, the GM and GSD are the two key descriptors. Truncation limits can also be specified, and correlations between parameters can be coded into the stochastic simulations. Sheppard and Sheppard (1989) discuss the impact of a stochastic correlation between CR and a measure of solubility of the element in the soil (K_d) on the decrease in the variation of model estimates.

Inclusivity of CR values

A major issue with the use of stochastic models is inclusivity: what range of conditions are considered appropriately in the PDF, and how important are outliers? For example, it could be argued that a PDF based on data for agronomic crops will not necessarily reflect soil-to-plant transfer for native plants. This becomes important for human cultures that consume large amounts of native plants or wild animals that consume such plants. Native plants differ in many respects from agronomic crops, most notably in potential growth rate, age at harvest, and in fertility of the substrate soil. The question of inclusivity will remain open until CR values have been obtained for a great many plants. However, some insight can be gained by considering the range of values now available, including the apparent outliers, and by considering the data we now have for native plants. The IUR database provides the best estimate available of the range of data for agronomic crops.

MATERIALS AND METHODS

Variation in CR values

The database was acquired for this study in 1995. The data were summarized and correlations among CR values and ancillary data were reported by Sheppard (1995). This report set out screening criteria for the data that were applied here as well. The screening criteria were stringently applied for elements with more than 100 CR values, but were relaxed for other elements to not unduly reduce sample size. Data for plants grown in pots or plants grown in soil with less than the required depth of contamination were considered for rejection. For cesium, plants sampled less than 1 y after contamination were rejected to avoid data that involved direct contamination from atmospheric releases such as Chernobyl. After screening, the overall GSD calculated for each element, without any categorization of the data (e.g., into categories for crop or soil type), was considered the fully generic GSD. We consider it appropriate for stochastic modeling where a specific site has not been chosen or assumed.

Sheppard (1995) also applied a statistical model to allow interpolation and limited extrapolation. The model provided estimates of CR for 10 to 20 crops for each element, adjusted to an arbitrary "standard soil" of pH 7, 15% clay and 5% organic matter content. Accompanying these estimates and derived from the same model were correction factors to adjust the CR values to represent soils ranging from pH 4 to 8, clay contents up to 60%, and organic matter contents up to 30%. These were analysis-of-variance statistical models, each used about 30 degrees of freedom (i.e., had about 31 combinations of factors specified in the model), each was statistically significant, and all are referred to here as detailed models. The residual sum of squares from these detailed models is used here to calculate GSDs for results of a detailed model. This would be the appropriate GSD when general soil types and crops are known, but there are no site-specific CR data.

The GSD for a full model was also calculated for each element by analysis of variance. This used all ancillary information in the database as independent variables. The full models were mixed models with both categorical variables (e.g., crop and soil types), and continuous variables (e.g., soil pH and organic matter content). These models used upward to 175 degrees of freedom (176 combinations of factors), and were taken to represent the most information that modelers might ever hope to obtain for practical assessments, apart from site-specific CR values. The GSD was the residual sum of squares and is largely for comparison only as few situations would have the large amount of independent information required by the model.

Intermediate models were also calculated, to show the decrease in GSD possible when model complexity increased. Obviously, some models will be more effective than others at decreasing the GSD for a set number of model degrees-of-freedom. However, the general relationship is of interest to examine the benefit of more detailed models.

Inclusivity of CR values

Inclusivity was examined by compiling CR values for native plants and comparing these to the PDF values

from the IUR database and from Zach and Sheppard (1991). This was done for cesium, where there are the most CR values, and for iodine, which is the most critical element in the disposal of nuclear fuel waste by the Canadian concept.

RESULTS AND DISCUSSION

Limitations of the database

A very large database with each value independent from all others would be required to fully investigate GSD values. Such a database is not available for all elements, but the IUR database is the best available. The limitations are important to recognize. For most of the elements with over 300 CR values, there were many data contributors and many conditions are represented. For cesium, the 1,115 CR values after screening came from 544 different combinations of soil pH, clay content, and organic matter content. These could be considered quite independent. In contrast, the 457 CR values for neptunium came from only 14 different combinations of soil pH, clay content and organic matter content and were apparently provided by only two investigators, so that independence is limited accordingly.

Dependence of CR on soil and crop variables

Because there are more data for cesium than for the other elements, they were used to investigate model complexity. Among all the ancillary variables, crop type explained the most variation in CR values (Sheppard 1995). The more detailed the description of crop type (i.e., the more degrees of freedom used), the more variation that was explained, and because of the very large total number of degrees of freedom for cesium, most of these models were highly significant. Sheppard (1995) limited the crop descriptions in the detailed models to less than 20 degrees of freedom, only because users would seldom be able to specify crop details (e.g., forage crop species) to greater precision. Continuous variables also significantly affected CR, even though all the correlation coefficients (r) were numerically small. For example, the r for soil pH was -0.25 ($p < 0.0001$) and for log of organic matter content it was $+0.19$ ($p < 0.001$). Scattergrams in Fig. 1 show these relationships. These results indicate that CR values are related to site-specific conditions, but there is still a large amount of variability.

Variation in CR

It might be expected that GSD values would be most inconsistent when there were very few underlying CR values. The GSDs may converge as the number of CR values increase. This trend is not evident for the IUR data (Fig. 2), except perhaps for cerium where there were only seven values. There are other points to note. Both manganese and zinc are plant essential elements, and they have low GSDs compared to other non-essential elements with about the same number of CR values (Fig. 2). This might be expected since manganese and zinc are more likely to be physiologically regulated than the others, and the stable isotopes are bioavailable in soil. The high GSD for technetium may reflect two things: 1) technetium undergoes very large changes in bioavailability with redox conditions and these were not recorded in the database, and 2) in the oxidized state technetium is so bioavailable that plants may quantitatively deplete it from the soil, and some researchers may not have noticed the extent of the depletion.

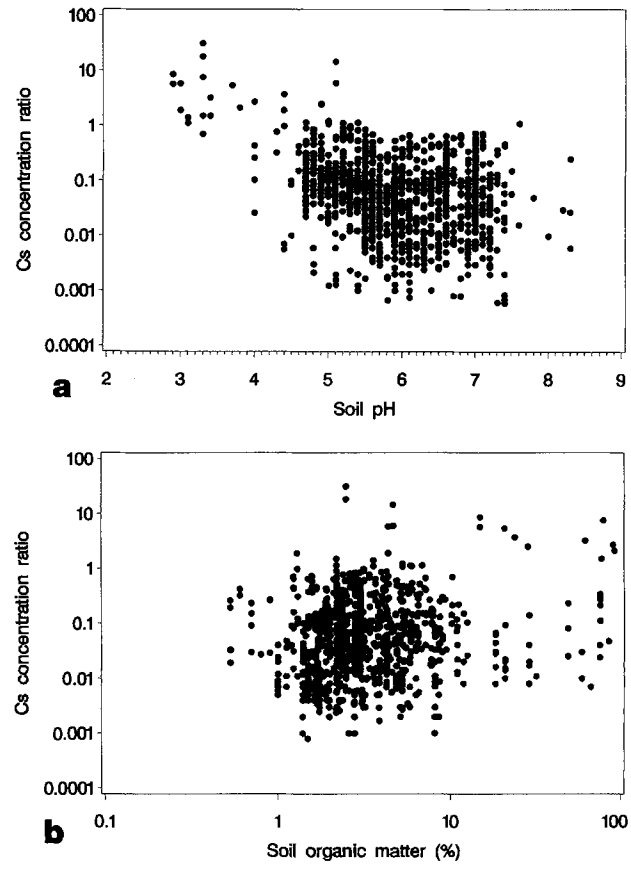

Fig. 1. Scattergrams of CR values for cesium as a function of soil pH (a) and organic matter content (b), with 1,115 values shown.

The IUR database has values for (arithmetic) standard deviations (SDs) associated with a total of 556 CR values. These SDs represent within-study, and often within-site, variation. Although there is no exact way to compare SD with GSD, one plus the coefficient of variation (1 + SD/mean) is an approximation of the corresponding GSD. There were some GSDs recorded as SDs in the database, all from one investigator, and these were used directly. For the 17 elements where coefficients of variation could be computed, the approximated GSDs ranged from 1.01 to 2.3 (Fig. 2) and were not correlated to the across-site measures of GSD for the elements. The estimates based on SDs represent the least spatial and temporal variation, and therefore the lowest GSDs possible. They might be typical of values that would be obtained if CRs were determined at the specific site being assessed.

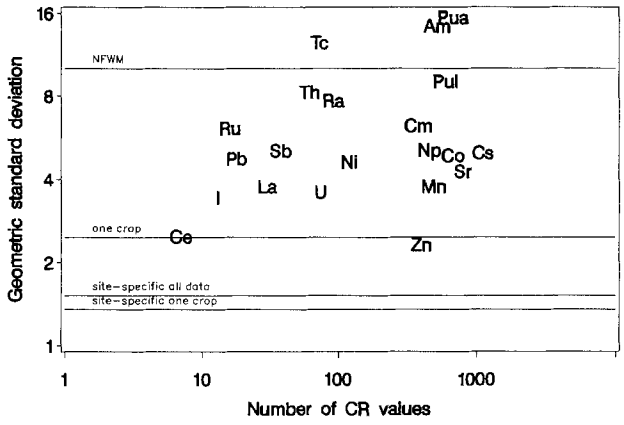

Fig. 2. Fully generic GSD values for 20 elements vs. the number of CR values for each element. Plotting symbols are the element symbols, except that Pul is plutonium from lab settings and Pua is plutonium from all settings. The horizontal lines are GSD values for the fully generic model of Zach and Sheppard (1991); one crop and 23 elements from Sheppard and Evenden (1990); site-specific CR values from the IUR database; and site-specific CR values from Sheppard and Evenden (1990).

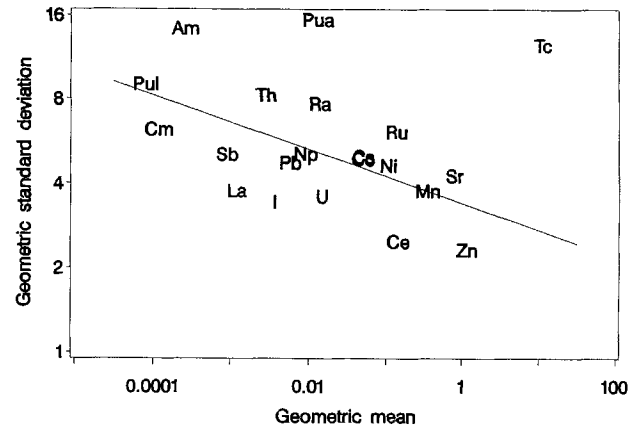

Fig. 3. Relationship of GSD to GM for 20 elements. Plotting symbols are the element symbols as before, and cesium and cobalt overlap. The line is the regression line $\log_{10}\text{GSD} = 0.53 - 0.096(\log_{10}\text{GM})$, where both coefficients are statistically significant ($p < 0.005$).

Sheppard and Evenden (1990) obtained a similar relationship between geographically generic and site-specific GSD values. They measured CR for 23 elements across 64 sites in one plant species (blueberry). The GSD for the survey, pooled across elements, was 2.47, but for 16 samples at one site it was 1.36. It is apparent from the IUR data that most site-specific data will have a GSD of about 1.5, and when data are fully generic the GSD will generally be above 3. The pooled GSD from the survey by Sheppard and Evenden (1990) of 2.47 may be appropriate for the situation where data are available for a specified crop, but not for a specified site.

The GSD chosen for all elements by Zach and Sheppard (1991) was 10, and this value was used in stochastic assessments. It was chosen so that the PDFs would be inclusive of settings and plant types not previously considered. Although based on scant information at the time, it is clearly an upper GSD value when compared to the IUR data (Fig. 2). Higher GSD values may be considered conservative in dose assessments because they increase the upper fractile dose estimates.

It is most probable that GSD values vary among elements, even though for lack of better information Zach and Sheppard (1991) used a single value. The example of plant-essential elements was already discussed. Relatedly, the IUR data show, with the exception of technetium, a distinct negative relationship between GSD and GM (Fig. 3): GSD values are larger when the GM is smaller. The reason for this is not clear, but it may relate to measurement. When CR is very small, such as for plutonium, the plant concentration is very low compared to the soil concentration and the potential for cross contamination is very large. Washing the plant may not be sufficient to remove all adhering soil. Soil adhesion introduces a new transfer pathway for elements with low CR values, and this added pathway, whose impact will vary among studies, may cause the apparent increase in GSD. The IUR database included a binary variable for washing, but the washing practices may not have been sufficient and there may have been undetected soil adhesion (Sheppard and Evenden 1995).

Contrary to the overall pattern in Fig. 3, the GSD for technetium was high despite the high CR. A possible reason for this, related to quantitative uptake of technetium from soil by plants, was already advanced.

Decrease in GSD with increase in model detail

The fully generic GSD encompasses variation resulting from spatial and temporal effects, crop and climatic effects, experimental variation and artifacts, and other unknown or unknowable sources. The GSD as residual error from the full model is the variation unexplained after all recorded information has been accounted. This should be as low a GSD as possible without site-specific CR data. The intermediate models vary in effectiveness, shown for cesium in Fig. 4, but it is apparent that even a modest model with few degrees of freedom is beneficial, while complicated models do not provide a lot of additional reduction in residual GSD. Based on arguments throughout this paper, appropriate GSD values are proposed for varying levels of information (Table 1).

McGee et al. (1996) make the case that ratios such as CR are not appropriate values to use in radioecology. They observed that CR for cesium decreased with increases in soil cesium concentration, as was reported earlier by Sheppard and Evenden (1988a) for uranium, thorium and lead. This implies a non-relationship or a non-linear relationship between plant radionuclide concentration and soil radionuclide concentration, a point that has also been made (Simon and Ibrahim 1987; Sheppard and Evenden 1988b, 1990). We regard non-linearity as one of the many sources of variability in CR

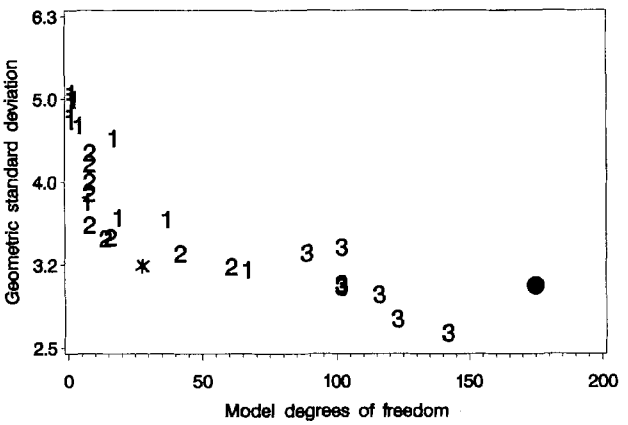

Fig. 4. The values of GSD for cesium given varying amounts of information, indicated by model degrees-of-freedom (df). The fully generic GSD has 0 df and the model including all recorded information (shown by a dot) had 175 df. The numerical plot symbols indicate the number of factors in each model, for example crop type and soil type would comprise a model with two factors and 61 df. The star indicates the detailed model of Sheppard (1995).

Table 1. Geometric standard deviations (GSDs) of the soil-to-plant concentration ratio (CR) appropriate to varying levels of information about the site and crops under investigation.

Level of information about site and crop	Appropriate GSD[a]
No information about site or crops: fully generic CR values	5.7 (2.4–16)
Soil type specified but no information about possible crops	4.4 (2.3–10)
Crop type specified but no information about site or soils	3.3 (1.8–7.4)
Limited information about both crops and soils, such as with the detailed models of Sheppard (1995)	3.2[b]
Extensive information about the site and crops, but no CR data from the site: full models from the IUR database	3.2 (1.9–5.1)
Site- and crop-specific CR values obtained on the site of interest	1.5 (1.1–3.7)

[a] The average GSD for 16 to 19 elements shown, with the range in brackets.
[b] Detailed models were developed for only four elements; this is an estimate for a more complete suite of elements.

values, and non-relationship is usually reported when only narrow ranges in soil concentration were studied. Clearly, based on the GSDs in Table 1, soil concentrations must range several orders of magnitude before a statistically significant relationship between plant and soil concentrations would be expected. The CR model still has merit, although as we said previously (Sheppard and Evenden 1990), it may only be to "effectively differentiate the behavior of the different elements." It is interesting to note that the overall variation in CR observed by McGee et al. (1996), across sites and species, had a sixfold range, consistent with the fully generic GSD in Table 1.

Inclusivity of CR values

The IUR database includes almost entirely data from agricultural or horticultural settings. These do not represent the native plants, and thus may be inappropriate when applied to natural or semi-natural settings. In addition, even among agronomic crops there is information lacking, especially given the recent diversification of crops to include exotics. As a result, it has become important to demonstrate the inclusivity, or lack thereof, of CR values. Our approach was to concentrate on two elements, cesium and iodine, and to search for unusual plants or settings, especially those with exceptionally high CR values. The exceptional data collected for cesium were obtained from non-agricultural crops listed in the IUR database, and from Bakken and Olsen (1990), Block (1990), Brown (1990), Caput et al. (1990), Colgan et al. (1990), Giovani et al. (1990), Horrill et al. (1990), Hove et al. (1994), Kopp et al. (1990), Maubert et al. (1990), Olsen (1994), Zach et al. (1989), and Zdenek and Benada (1990). Only CR values above 0.3 were collected for cesium, because of the vast amount of data available. Very high CR values for cesium resulting from direct contamination from Chernobyl were not used because they may represent adhesion to foliage rather than root uptake. Data for higher plants and for fungi (mushrooms) were kept and summarized separately. Because there are far fewer data in the literature for iodine than cesium, and because the IUR database does not represent iodine well, our compilation of CR values for iodine was not limited to non-agricultural or exceptional data. The data were obtained from Cline and Klepper (1975), Fukuzaki and Moriyama (1985), Haisch and Schüttelkopf (1993), Johnson and Butler (1957), Klepper et al. (1976), McGrath and Fleming (1988), Ohmomo et al. (1993), Pel and Schüttelkopf (1995), Robens et al. (1988), Schmitz and Aumann (1994), Sheppard et al. (1993), Sheppard and Evenden (1988b), Sheppard and Motycka (1997) Tikhomirov and Rusina (1983), Whitehead (1975), Wildung et al. (1985) and Yuita (1994).

Fig. 5 shows the probability distributions for the fully generic IUR data, that for the Canadian NFWM program, and the actual frequency for selected data from the literature. For cesium, the selected data were of non-agricultural plants, and since they are on or above the upper tail of the IUR distribution it is evident that the IUR distribution does not represent non-agricultural plants well. However, non-agricultural plants are at least partially represented by the broad distribution used in the NFWM program. The distribution for the NFWM program allowed for 34% of the CR values for cesium to be above 0.3, in the range we recorded as exceptional and non-agricultural. The data for fungi are above the NFWM distribution, only 1% of the distribution is in the range of the data for fungi. Fungi are clearly a special case, especially for cesium, and the CR approach may not be appropriate.

The histograms for iodine in Fig. 5 show all the data we found in the literature, not just non-agricultural plants. The histogram agrees very well with the distribu-

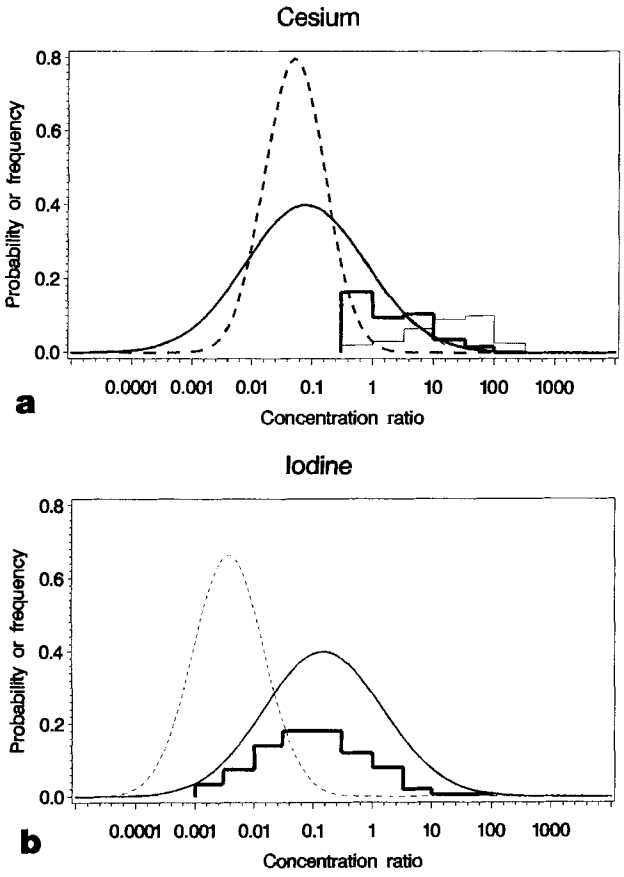

Fig. 5. Probability distributions of CR values for cesium (a) and iodine (b), showing with a solid-line curve the fully generic distribution from Zach and Sheppard (1991) and with the dashed-line curve the distribution of the IUR data with the overall GM and a GSD corresponding to a detailed model. The frequency histograms in bold lines are of data from references identified in the text (the frequencies are divided by 200 to fit the probability scale). For cesium, CR values for fungi are shown by the histogram demarcated with fine lines.

tion used for the NFWM program but is above that of the IUR. The IUR data are for grass only. Some of the highest values for iodine were for wetland crops such as rice (Sheppard and Motycka 1997).

CONCLUSION

The variation in CR values can be characterized by GSDs, and these are applicable across elements and settings. The best measures of GSD come from very large databases, and fortunately these GSDs can be used for elements where there are fewer data. The GSD appropriate for fully generic, stochastic use of CR is about 6. With more information about the site being assessed, the GSD may decrease to about 3.2. With site-specific CR data, the GSD is still about 1.5. More attention in the literature to the values of GSD for stochastic assessments will improve these estimates further, perhaps to even allow element-specific estimates.

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