

Research Article

Comparison of Methodologies to Estimate Intake Dose for Exposure to Soil Contaminants

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It has been suggested that probabilistic approaches would provide more realistic estimates for human intake dose from exposure to soil contaminants than the commonly-used standard deterministic method. The objective of this study was to compare intake dose estimated by these methods for noncarcinogens and carcinogens in soil from 21 contaminated sites in Pennsylvania, USA. Intake doses by the principal human exposure routes for these contaminants were estimated by the standard deterministic method using fixed input parameter values, and by two emergent probabilistic methods. The probabilistic methods were based (a) on distribution functions for all input parameters, or (b) on some combination of these functions and fixed parameter values. Intake doses were then taken as the 90th, 95th, or 99.9th percentile of the generated cumulative output distribution and compared with the commonly-used deterministic estimates over all contaminant/site combinations. For all exposure routes, the 90th and 95th percentile intake dose estimates were not markedly different from the deterministic values or from each other. The opposite was generally the case for the 99.9th percentile estimates. These results did not indicate clear and definitive advantages in using probabilistic methods over the deterministic method for estimating human intake dose from exposure to soil contaminants.

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1. Introduction

In industrialized countries with strong enforcement of environmental laws and regulations, many manufacturing facilities have been abandoned or idled because of real or perceived soil contamination. These facilities are usually in urban areas, and costly governmental and private programs have sought to stimulate their redevelopment in order to regenerate jobs that have been lost due to their closure. Facilities with economical potential for redevelopment are called brownfields [1]. The soil contamination associated with brownfields creates legal liabilities for potential redevelopers. This liability can eliminate the possibility of obtaining bank loans for the redevelopment of the facility. The financial cost and risk of investing in brownfields are therefore a direct function of the risk for deleterious health effects to the surrounding community from potential exposure to contaminants at the site [1]. The first step in assessing this risk is to estimate intake dose by the principal human exposure routes for contaminants present at the brownfield.

The intake dose is then combined with established toxicity values to determine the human health risk for a given soil contaminant. Greater health risk will mean higher costs of soil cleanup and lower likelihood of reinvestment in these properties [1].

The algorithms for estimating intake dose require data on the type and concentration of the contaminant together with many exposure input parameters [1]. Currently, most algorithms use some fixed upper-percentile value (usually the 95th percentile) obtained from statistical analysis of the observed concentrations for a given soil contaminant, together with fixed standard recommended values for exposure input parameters such as intake rates, exposure time, exposure duration, and body weight [2]. To obviate any possibility of endangering public health and welfare the fixed input parameters are often chosen as the maximum (or minimum as appropriate) over the range of possible values to ensure “erring on the side of safety” [3]. This upper-percentile level of contamination and these fixed exposure input parameters are then used in the algorithms

to determine a single estimate of the intake dose. This approach to estimating the intake dose for human health risk assessment is termed as deterministic.

Variability and uncertainty are always associated not only with the observed contaminant levels but also with the exposure input parameters and toxicity values, and these are not accounted for in the deterministic risk assessment approach [2]. Bogen [4], Burmaster and Anderson [5], and USEPA [6] have strongly suggested that the deterministic approach overestimates the intake dose, and that probabilistic-based approaches would provide more realistic estimates.

Probabilistic-based methodologies use probability density distribution functions for the observed contaminant levels and exposure factors instead of fixed values. Unlike the deterministic approach that provides only a single estimate of the intake dose, probabilistic-based methodologies generate a cumulative distribution of intake doses that would better account for variability and uncertainty. An intake dose value in the 90th to 99.9th percentiles of this distribution is then used for further risk assessment [6]. A study by Smith [7] indicated that the deterministic approach would give similar intake dose estimates as the 95th percentile of the intake dose distribution obtained by such a probabilistic-based approach.

In the probabilistic-based approach, the cumulative intake dose distribution is generated using Monte Carlo simulations. Once the appropriate probability distribution functions for the input variables have been developed, values are selected at random from these distributions. These are then used as input into the algorithms to calculate a single value of the intake dose as is done for the deterministic approach [8]. This process is then repeated thousands of times to develop a cumulative distribution of intake doses.

This Monte Carlo-based probabilistic methodology allows for sensitivity analysis to determine which input parameters would have the least impact on the intake dose estimate of a simulation. Pearson's r or Spearman's ρ can be used to test for sensitivity of the intake dose estimates to each parameter [9, 10]. The nonsignificant input parameters can then be treated as deterministic variables. This hybrid methodology, between the purely Monte Carlo and deterministic approaches, reduces the number of distributions that must be developed and the number of Monte Carlo simulations that must be performed.

Clearly the need exists to investigate the applicability and advantage of using Monte Carlo-based probabilistic methodologies to better capture variability and uncertainty in brownfields risk assessments vis-à-vis the standard deterministic approach. It is possible that the Monte Carlo-based methodologies may not produce significantly different intake dose estimates compared to the deterministic method. However, if it can be shown that Monte Carlo-based risk assessments can better capture the uncertainty and variability inherent to the risk assessment process, it will have far-reaching financial implications for brownfield cleanup and redevelopment.

The primary objective of this study was to compare intake dose estimates for the principal human exposure routes obtained by the deterministic and Monte Carlo-based

probabilistic methodologies using measured soil levels of noncarcinogens and carcinogens at a number of randomly-chosen brownfield sites.

2. Materials and Methods

A reliable set of brownfield contaminant data was developed using the Pennsylvania Department of Environmental Protection (PADEP) Land Recycling Program list of brownfields for which site investigations were completed. The list at the end of 2000 contained 633 such completed brownfields. These 633 sites were sorted into four groups based on whether the site was closed to background, statewide generic standards, site-specific, or industrial standards. Only the sites that were closed to statewide health, site-specific, and industrial standards were used in this study. From this reduced list, 30 sites were selected at random. A formal request was made to PADEP to gain full access to the files on these 30 sites. Ten of these sites did not have sufficient data or the PADEP could not find the files. One site (Exxon SGH Specialty Products) was listed as one facility but had two separate site investigations. Each of these was treated separately, giving a total of 21 brownfield datasets used in this study.

Measured contaminant levels in the soil at these sites were used to estimate exposure concentrations in the air and groundwater. The concentrations were calculated using the procedures provided in the "Soil Screening Guidance: Technical Background Document" [11]. These procedures use several assumptions that simplify the underlying soil/air and soil/water mass transfer models [11] and reduce the need for large quantities of site-specific data that are often not available.

The calculation of exposure concentrations for the inhalation route is divided into two possible mechanisms: inhalation of vapors for volatile contaminants and inhalation of contaminants adsorbed onto suspended soil particles for nonvolatile contaminants. The concentration of contaminant in the air for volatile and nonvolatile contaminants is calculated using the volatilization factor (VF) or the particulate emission factor (PEF), respectively. Both the VF and PEF are based on two models: one that estimates vapor or particulate emissions from the soil and one that estimates subsequent atmospheric dispersion [11].

For the deterministic approach, the single upper-percentile soil, air, and groundwater exposure concentrations were calculated using Land's method as outlined "Supplemental Guidance to RAGS: Calculating the Concentration Term" [12]. Land's formula uses the mean (\bar{x}) and standard deviation (s_x) of n log-transformed sample data points to calculate the 95th percentile upper confidence level ($UCL_{0.95}$) of the sample data as [12]

$$UCL_{0.95} = \exp \left[\bar{x} + s_x + \frac{s_x h_{0.95}}{\sqrt{n-1}} \right], \quad (1)$$

where $h_{0.95}$ is the h -statistic for confidence level 0.95 [12]. The h -statistic is a unique symmetric unbiased estimator for the central moment of a distribution [13].

When there is high standard deviation or low sample size, Land's formula provides extremely high estimates for the $UCL_{0.95}$. In those cases, the maximum detected or modeled concentration was used for the upper percentile value [12]. Where the value of the sample concentration was reported as nondetect or below the detection limit, the sample was treated as coming from a noncontaminated area and was removed from further consideration [14].

The deterministic intake dose estimates for individual contaminants were calculated in accordance with the procedures outlined in "Risk Assessment Guidance for Superfund Volume I: Human Health Evaluation Manual (Part A)" Chapters 6, 7, and 8 [2]. Exposure parameters were taken from "The Exposure Assessment Handbook" and "USEPA Region III Risk-Based Concentration Table: Technical Background Information" [15].

The Monte Carlo-based and hybrid Monte Carlo/deterministic-based intake dose distributions for individual contaminants were calculated using @Risk 4.0, Advanced Risk Analysis for Spreadsheets (Palisade Corporation, Newfield, NY, USA), a Monte Carlo simulator that is a plug-in to EXCEL (Microsoft Corporation, Redmond, Wash, USA). Recommended probability density functions for the input parameters were obtained from the "Support Document for the Development of Generic Numerical Standards and Risk Assessment Procedures" issued by the Ohio Environmental Protection Agency (Columbus, Ohio 43215, USA) [16], and from Finley et al. [17]. @Risk 4.0 calls specific function routines to generate the type of distribution specified for a given input parameter to be used in the Monte-Carlo simulation.

The @Risk 4.0 best fit function was used to fit probability density functions to the observed soil concentration data values. The goodness of fit was determined using the Chi-Squared Statistic, the Kolmogorov-Smirnov statistic, and the Anderson-Darling statistic. Only the normal, lognormal, uniform, and triangular distributions were fitted to the soil concentration data. Ten thousand iterations of the Monte Carlo and Monte Carlo/deterministic simulations were run using Latin hypercube sampling to ensure convergence for the tails of the output distribution of intake doses. The results were then used to calculate the 90th, 95th, and 99.9th percentiles of the intake dose distribution.

Sensitivity analysis was performed using the Monte Carlo simulations to determine which of the parameters could be treated as deterministic. The sensitivity analysis tool in @Risk 4.0 was used to perform the sensitivity analysis. Multivariate stepwise regression was used to determine the R -squared values. The parameters with the lowest R -squared values were then treated as deterministic values in the Monte Carlo/deterministic simulations.

Seven sets of intake doses were estimated and compared. These were the deterministic estimates and the 90th, 95th, and 99.9th percentiles of the estimated output distributions from the Monte Carlo and the Monte-Carlo/deterministic methods. These estimated intake dose levels were laid out according to a randomized complete block design where each contaminant/site combination was a block and each estimating method was a treatment. A one-way analysis of

variance (ANOVA) was performed for this design and the treatment means of the intake dose levels were compared for significant differences using Tukey's LSD. Other designs with 2-way ANOVA for the intake dose estimates were possible. However, analyzing the intake dose estimates in this manner allowed us (a) to examine overall trends in the relative performance of the 7 estimation methods over varying contaminants, sites, and exposure routes, and (b) to use the mean intake dose averaged across all contaminant/site combinations for each method as a single response variable for comparison of the 7 methods. All statistical analyses were performed using SAS (SAS Institute, Cary, NC 27513, USA).

Analyses of variance were performed for estimated noncarcinogenic intake dose levels corresponding to 69 contaminant/site combinations for the soil ingestion, soil dermal absorption, and groundwater ingestion exposure routes as listed in Table 1. Analysis of the noncarcinogenic inhalation intake dose levels was done for 45 contaminant/site combinations as listed in Table 1.

Intake dose levels of carcinogens for 30 contaminant/site combinations were estimated for the soil ingestion, dermal absorption, and groundwater ingestion exposure routes and were analyzed as for the noncarcinogens. These are listed in Table 2.

It should be noted that since USEPA lists methylene chloride to be only a probable human carcinogen it was treated as both a noncarcinogen and carcinogen and appears as a contaminant in both Tables 1 and 2.

3. Results and Discussion

For convenience, in the following discussion we designated the intake dose estimation methods as deterministic (DET), the Monte Carlo probabilistic (MC), and the Monte Carlo probabilistic/deterministic (MCD). In addition, we attached 90, 95, or 99.9 to MC or MCD as appropriate to indicate that the intake dose for each contaminant/site combination using the probabilistic methods was taken as the 90th, 95th or the 99.9th percentile of the estimated cumulative intake dose distribution. The mean intake dose (MID) and the standard error of the mean resulting from the analysis of variance were used to demonstrate differences between the 7 intake dose estimation methodologies for the noncarcinogens and carcinogens by each human exposure route (see Tables 1 and 2).

Figure 1 shows the MID by the soil ingestion exposure route for noncarcinogens. The deterministic MID estimate was not significantly different from those for the MC 90 and MC 95 or the MCD 90 and MCD 95 treatments. However, the 99.9th percentile MID estimates for the MC and MCD methods were significantly different from the deterministic value. For all percentile levels of the MCD method, there were no significant differences in the MID estimates as compared to the corresponding MC generated values. In summary, inclusion of deterministic values in the MCD estimation of the intake dose did not significantly influence the outcome compared to the MC method.

Figure 2 shows the MID estimates for the dermal absorption exposure route. The MID estimates by this exposure

TABLE 1: Contaminant/site combinations and exposure routes for which intake dose for noncarcinogens was estimated.

Contaminant	No. of sites	Soil ingestion	Dermal absorption	Inhalation	Groundwater ingestion
Naphthalene	10	X	X	X	X
Ethylbenzene	7	X	X	X	X
Methylene chloride	7	X	X	X	X
Phenathrene	7	X	X	—	X
Toluene	6	X	X	X	X
Arsenic	6	X	X	—	X
Acetone	5	X	X	X	X
Barium	5	X	X	X	X
Chromium	5	X	X	X	X
Fluoranthene	5	X	X	—	X
Xylene	5	X	X	—	X
Totals	69	69	69	45	69

TABLE 2: Contaminant/site combinations and exposure routes for which intake dose for carcinogens was estimated.

Contaminant	No. of sites	Soil ingestion	Dermal absorption	Groundwater ingestion
Arsenic	6	X	X	X
Benzo[a]anthracene	6	X	X	X
Benzo[a]pyrene	6	X	X	X
Chrysene	5	X	X	X
Methylene chloride	7	X	X	X
Totals	30	30	30	30

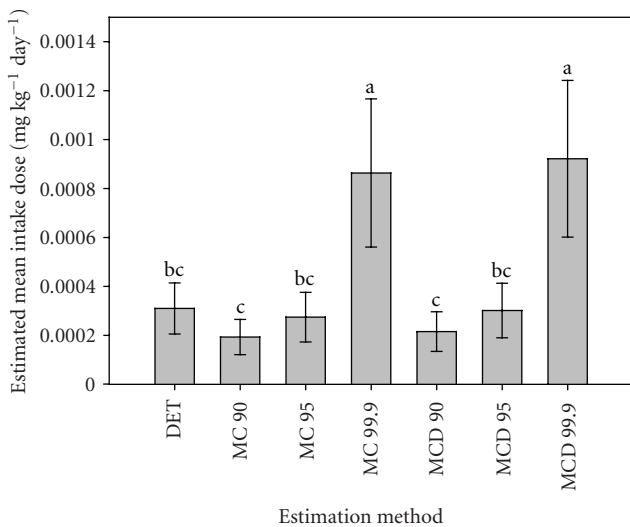


FIGURE 1: Estimated mean intake dose (MID) by the soil ingestion exposure route for the noncarcinogens. Values not annotated with the same letter are significantly different at $P \leq .05$. Bars represent the standard error of the mean.

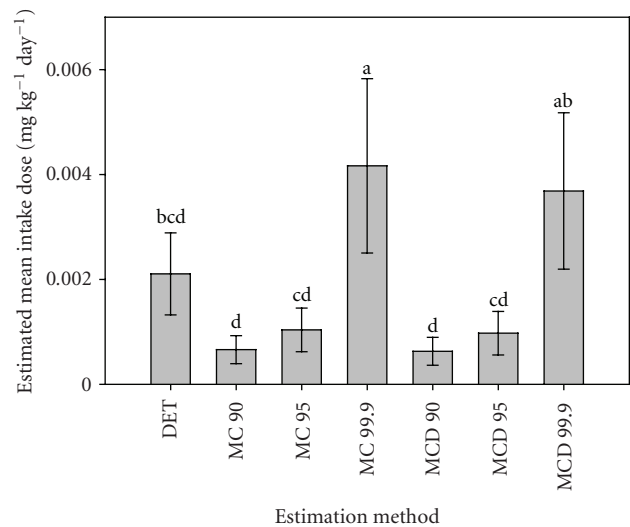


FIGURE 2: Estimated mean intake dose (MID) by the dermal exposure route for the noncarcinogens. Values not annotated with the same letter are significantly different at $P \leq .05$. Bars represent the standard error of the mean.

route are an order of magnitude greater than those for the direct soil ingestion route. The pattern of the response to the various estimation methods is very similar to that in Figure 1. The MID estimate calculated using the deterministic method was not significantly different from those generated by the MC 90 and MC 95 or the MCD method at the 90th, 95th, and 99.9th percentiles. However, the MID estimate for the MC

99.9 method was significantly different from the value for the deterministic method. For all percentile levels of the MCD method, there were no significant differences in the MID estimates compared to the corresponding values generated by the MC method.

Figure 3 shows the MID estimate for the inhalation exposure route. These MID estimates were, respectively, 3 and 2

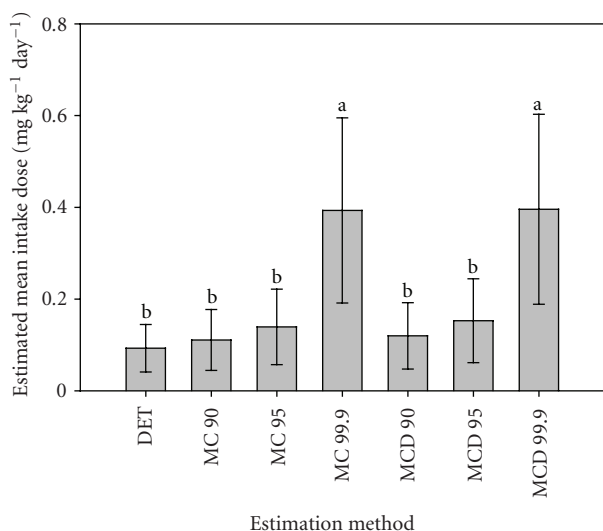


FIGURE 3: Estimated mean intake dose (MID) by the inhalation exposure route for the noncarcinogens. Values not annotated with the same letter are significantly different at $P \leq .05$. Bars represent the standard error of the mean.

orders of magnitude higher than those for noncarcinogens administered by the soil ingestion and dermal exposure routes. However, the pattern of the response of the MID estimates to the different methods is quite similar to those of the other two routes. The MID estimate calculated by the deterministic method was significantly different only when compared to the MC 99.9 and MCD 99.9 methods.

The MID estimates by the ingestion-of-groundwater exposure route for noncarcinogens are shown in Figure 4. These values were almost the same as for the inhalation exposure route except that the MID estimate of the deterministic method was 4 times greater. As for the inhalation exposure route, the deterministic MID estimate was significantly different from those generated by the MC and the MCD methods at the 90th and 95th but not for the 99.9th percentiles. These latter MID estimates were not significantly different from each other.

Regardless of the exposure route, the MID estimates for noncarcinogens showed a similar response pattern for the different estimation methods. As would be expected, the MID estimates for all the probabilistic methods increased nonlinearly as the cutoff limits on the output probability distribution for the intake dose were raised from the 90th to the 95th and 99.9th percentiles. For all the exposure routes, the MC 99.9 and MCD 99.9 methods gave the highest MID estimates.

Figure 5 shows the MID estimates for the carcinogens by the soil ingestion exposure route. Similar results are presented in Figures 6 and 7 for the dermal and groundwater ingestion exposure routes. As pointed out earlier, results for the inhalation exposure route for carcinogens were not generated since the available data set did not contain enough contaminant/site combinations and toxicity values for carcinogens were unavailable to adequately perform the analyses. As shown in Figures 5 to 7, the MID estimates

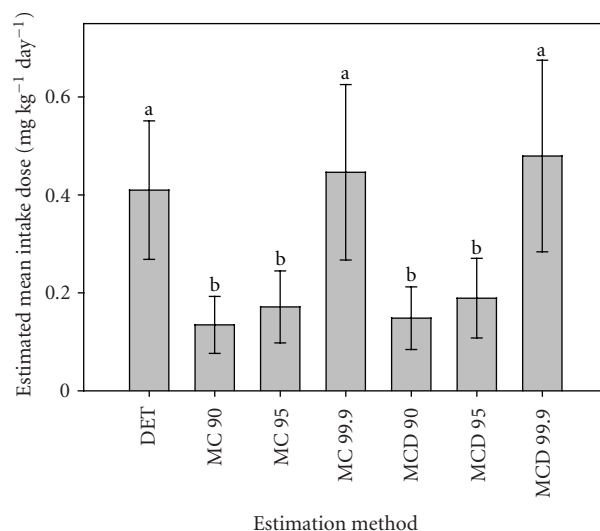


FIGURE 4: Estimated mean intake dose (MID) by the ingestion-of-groundwater exposure route for the noncarcinogens. Values not annotated with the same letter are significantly different at $P \leq .05$. Bars represent the standard error of the mean.

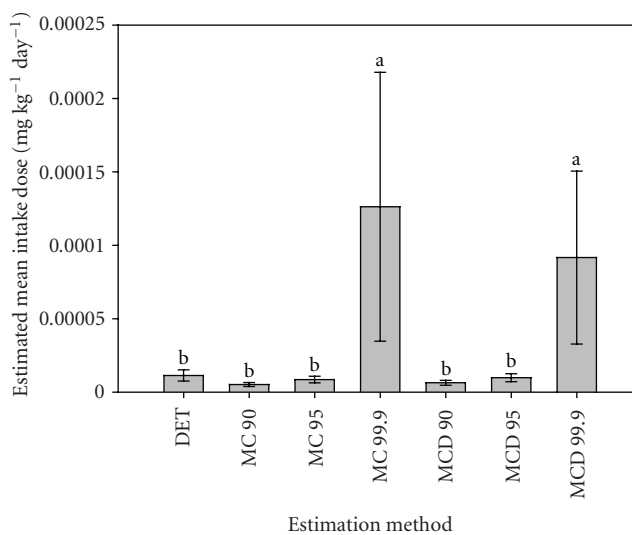


FIGURE 5: Estimated mean intake dose (MID) by the soil ingestion exposure route for the carcinogens. Values not annotated with the same letter are significantly different at $P \leq .05$. Bars represent the standard error of the mean.

are 1 or 2 orders of magnitude lower than those for noncarcinogens for a given exposure route. This is because the measured levels for carcinogens were relatively lower than those for noncarcinogens at all the 21 brownfields sites. Additionally, for carcinogens, the averaging time is taken as 70 years, which is greater than the exposure duration. For noncarcinogens, it is the same as the exposure duration.

The MID estimates for carcinogens by the various methods follow the overall pattern shown in the results for noncarcinogens. The highest MID estimates were obtained with the MC 99.9 and MCD 99.9 methods, although, unlike

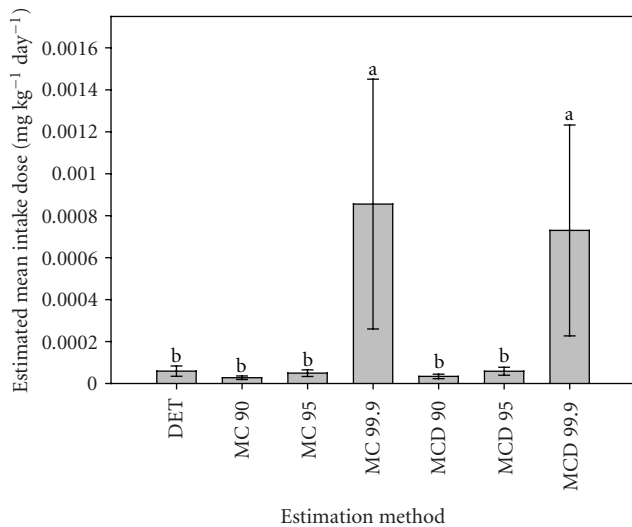


FIGURE 6: Estimated mean intake dose (MID) by the dermal exposure route for the carcinogens. Values not annotated with the same letter are significantly different at $P \leq .05$. Bars represent the standard error of the mean.

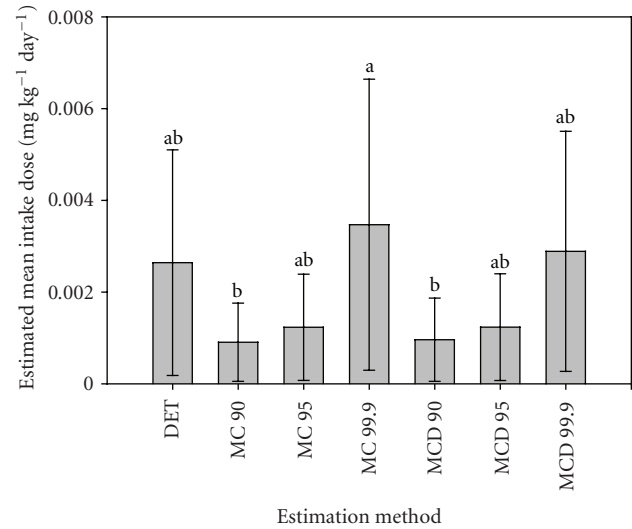


FIGURE 7: Estimated mean intake dose (MID) by the ingestion-of-groundwater exposure route for the carcinogens. Values not annotated with the same letter are significantly different at $P \leq .05$. Bars represent the standard error of the mean.

the results for noncarcinogens, the latter were consistently lower than the former. As for noncarcinogens, the MID estimates for the carcinogens increased nonlinearly as the cutoff limits on the output intake dose distribution were raised to the higher percentiles. However, as shown in Figures 5 and 6, these increases were much steeper than those for noncarcinogens except for the groundwater ingestion route (see Figure 7). This would indicate that the tails of the output intake dose distributions by the MC and MCD methods were much steeper than the corresponding ones for the noncarcinogens. This is probably caused by the lower variability in the measured concentrations for the carcinogens.

Taken together, the results presented in Figures 1 through 7 were consistent with the findings of Smith [7]. The MID estimates obtained by the deterministic method for carcinogens and noncarcinogens for each exposure route were generally little different from those generated by the probabilistic methods except at the 99.9th percentile cutoff on the output intake dose distribution. We observed that several, but not all, of the fixed input parameter values used in the deterministic computations fell in the upper tails of their probability density distributions used in the Monte-Carlo simulations. Therefore, this finding would not appear to support the idea introduced by Bogen [4] that intake dose is overestimated if many upper-bound values of the input parameters are used in the deterministic method. On the other hand, other input parameter values that were closer to the mean of the distribution (such as body weight) would tend to offset such possible overestimation of the intake dose using the deterministic approach.

The results in Figures 1 through 7 clearly show that for both carcinogens and noncarcinogens the MC and the MCD methods would tend to produce the same intake doses

and risk estimates regardless of the exposure route. This is not surprising since using deterministic values in the Monte Carlo approach for the least sensitive input variables, as determined by sensitivity analyses, would not be expected to significantly impact the generated cumulative intake dose distribution.

4. Summary and Conclusion

Variability and uncertainty are intrinsic to each input parameter of the intake dose algorithms used for estimating human health risk from exposure to soil contaminants [3, 6, 8, 18]. Using fixed input parameters that represent maximum (or minimum) values over their range to calculate intake dose is the most common approach to “erring on the side of safety” in environmental exposure assessment [3]. The deterministic procedure is simple, accessible, and computationally straightforward [6]. In addition, the computation can be done in reverse to provide soil concentration levels for a specified exposure dose or risk, and used to screen site contaminant levels to decide if they should be considered for further investigation [18]. On the other hand, it has been suggested that using many extreme values could result in inflated and unrealistic exposure dose estimates [4, 18, 19].

Probabilistic exposure assessment makes use of the same basic algorithms but makes use of probability distribution of the input parameters instead of fixed values. This makes possible generating an output cumulative probability distribution of intake doses. Monte Carlo techniques are most frequently used to develop the input parameter probability distributions [18]. However, Monte Carlo-based exposure assessment is computationally more complicated and time consuming requiring thousands of iterations to stabilize the tails of the output exposure probability distribution. This

makes quality control and calculation checking more difficult than for the deterministic procedure. These drawbacks of the Monte Carlo-based approach can be partially overcome by applying sensitivity analysis to reduce the number of input probability distributions if their simulated impact on the output distribution is found to be inconsequential.

The results of this study in their entirety showed that use of Monte Carlo-based probabilistic methods to estimate intake dose at the low to moderate cutoff levels (90th and 95th percentiles) of the output cumulative distribution would not provide significantly different estimates as compared to the traditional fixed-parameter method. Taken together, these results indicated that there might be little advantage to choose probabilistic methods over the deterministic method for intake dose estimation and risk assessment for brownfield redevelopment.

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