Model comparison for risk assessment: A case study of contaminated groundwater

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Abstract

Many environmental multimedia risk assessment models have been developed and widely used along with increasing sophistication of the risk assessment method. Despite of the considerable improvement, uncertainty remains a primary threat to the credibility of and users' confidence in the model-based risk assessments. In particular, it has been indicated that scenario and model uncertainty may affect significantly the assessment outcome. Furthermore, the uncertainty resulting from choosing different models has been shown more important than that caused by parameter uncertainty. Based on the relationship between exposure pathways and estimated risk results, this study develops a screening procedure to compare the relative suitability between potential multimedia models, which would facilitate the reduction of uncertainty due to model selection. MEPAS, MMSOILS, and CalTOX models, combined with Monte Carlo simulation, are applied to a realistic groundwater-contaminated site to demonstrate the process. It is also shown that the identification of important parameters and exposure pathways, and implicitly, the subsequent design of uncertainty reduction and risk management measures, would be better-formed.
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1. Introduction

In many countries, more and more sites with contaminated groundwater caused by improper handling or disposal of hazardous materials or wastes have been found. These sites may cause adverse effects on the environment and human health, and thus need to be evaluated as to whether and what remediation scheme should be applied. Health risk assessment is deemed as the most important tool for quantifying human health impact associated with pollutant-releasing activities and hence has become widely used as an aid in environmental decision-making processes (Maxwell and Kastenberg, 1999; Ma, 2002).

Health risk assessment involves identifying the potential of a risk source to introduce risk agents into the environment, estimating the amount of risk agents that come into contact with the human–environment boundaries, and quantifying the health consequence of exposure. Since the risk assessment paradigm was established in 1983 (NRC, 1983), the methodology of risk assessment has become more sophisticated. Compared to traditional generic, single-medium, and deterministic risk assessment methods, site-specific, multimedia, and stochastic risk assessment has become common practice. However,

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despite of the considerable improvement of the method, uncertainty remains a primary threat to the credibility of and hence users' confidence in the model-based risk assessments. Recent research on risk assessment has focused on uncertainty, because the uncertainty associated with parameter inaccuracy or variation, model simplification and inadequacy, and unsuited scenario designation often perplexes the users and decision makers in the process of risk assessment and management. Nevertheless, except research related to parameter uncertainty, the analysis of model and scenario uncertainty in the recent literature is still rare. Although the contribution of scenario and model uncertainty to overall uncertainty is usually assumed negligible or ignored, both types of uncertainty may affect significantly the outcome of risk assessment (Moschandreas and Karuchit, 2002). Some researches tried to quantify scenario and model uncertainty, and the outcome revealed that the total uncertainty including scenario, model and parameter uncertainty is three times greater than that considering only parameter uncertainty (Moschandreas and Karuchit, 2002). Although it has been indicated that scenario and model uncertainty are the important contributors to the total uncertainty, there are no practical measures as to how model and scenario uncertainty could be reduced.

Currently, many multimedia risk assessment models for implementing site-specific risk assessment have been developed, including MEPAS (Buck et al., 1995), MMSOILS (USEPA, 1996), CalTOX (Mckone, 1993a,b,c), 3MRA (USEPA, 2003), and TRIM (USEPA, 2002a,b), etc. One of these or related models will be adopted based on characteristics of different contaminated sites and scenario assumptions. But it is a difficult task for an inexperienced modelers to choose from so many environmental multimedia models (Del Re and Trevisan, 1995; Garen et al., 1999). Quite a few researches have performed model comparison to study the relationship between model differences and estimated results recently. Although model developers have tried hard to avoid uncertainty existing in the process of model construction, the differences in model design, environmental mechanism, mathematical formulations, and assumptions can result in difference of risk predictions by orders of magnitude (Laniak et al., 1997; Mills et al., 1997; Regens et al., 2002). Moreover, the uncertainty resulting from choosing different models has been shown more important than that caused by parameter uncertainty (Pollock et al., 2002). However, the finding that the results produced from different models may vary significantly does not provide suggestion as to which model is the best, except underlining the importance of understanding the limitations and assumptions of these multimedia models, and the compatibility between conceptual model and multimedia models to avoid the large errors introduced through use of improper models (Mills et al., 1997; Whelan et al., 1999; Regens et al., 2002).

Although model selection will cause considerable model uncertainty, very few studies explored the quantification and reduction of model uncertainty. Hertwich et al. (2000) even concluded that scenario and model uncertainty could be only exploratory and difficult to analyze quantitatively. At present the quantification of model uncertainty is generally conducted by way of calculating the range of outputs of different models (Moschandreas and Karuchit, 2002). Therefore, developing a screening procedure to compare the relative suitability between potential multimedia models would facilitate the reduction of uncertainty due to model selection by excluding unsuitable model. In other words, this study attempts to find out the relation between exposure pathways and estimated risk results and use this information to select the proper model through comparison of environmental multimedia models. A realistic groundwater contamination site is used as the case study.

2. Materials and methods

2.1. The contaminated-groundwater problem and conceptual model development

The important first step to implement risk assessment for a contaminated site is to develop the conceptual model of the site (Regens et al., 2002). The conceptual model is a descriptive model which uses available information to define all sources, types, and concentrations of contaminants, potentially contaminated media, potential exposure pathways, and final receptors (USEPA, 1989, 1991). According to the developed conceptual model, the exposure pathways in each multimedia model can be selected to correspond to the practical scenario.

The case study considered in this research is about a site, located in northern Taiwan, where it was discovered that the soil and groundwater was contaminated by chlorinated hydrocarbons, primarily trichloroethylene (TCE) and tetrachloroethylene (PCE). These chemicals were used as degreasing solvents by a local factory but improper handling and disposal of wasted solvents led to leakage and subsequent contamination of soil and groundwater.

Extensive resources and efforts have been spent in cleaning up the site. The soil cleanup has been considered complete, but little success has been reached for groundwater, due to dense-non-aqueous-phase-liquid's distinct physical characteristics that increase the difficulty of its identification and removal. At this point, the government needs to determine an appropriate course of management of groundwater contamination and subsequent feasibility of land use and development.

Risk assessment has been called forth to evaluate the problem and to facilitate the decision-making.

Based on the land use property and spatial pattern of contaminant concentration, the site is divided into four zones, each of which is assessed individually. This paper uses a case of one of the four zones: 80 000 m^2 of farmland with scattered residences. According to the observations of 10 monitoring wells in this zone, the mean and standard error of the contaminant concentrations are 0.000741 and 0.000095 mg l^{-1} for TCE, respectively, and 0.000931 and 0.000285 mg l^{-1} for PCE, respectively (Taiwan EPA, 2003). The soil is deemed free of contamination according to the site investigation, and the contamination source in this case study is the groundwater, which the residents use for domestic purposes and irrigation. The contaminated media includes domestic water, indoor air, irrigation soil, vegetables, crops, and farm animals. The exposure pathways include ingestion of drinking water, ingestion of shower water, ingestion of beef, ingestion of milk, ingestion of vegetables, ingestion of crop, ingestion of soil, dermal contact of soil, dermal contact of shower water, inhalation of shower air, and inhalation of indoor air. Fig. 1 depicts the conceptual site model that illustrates the environmental transfer processes between media and exposure pathways linking exposure media and exposure routes involved in this specified scenario.

2.2. Multimedia risk assessment models

Many multimedia risk assessment models have been developed for performing site-specific risk assessment. For the purpose of demonstration, MEPAS, MMSOILS, and CalTOX are chosen for model comparison because these models are widely used environmental multimedia models and suitable for groundwater-contaminated site assessment.

The scenario included in this case study is that the residents use groundwater for irrigation and domestic purposes. In this scenario, comparing each multimedia model with the conceptual model can find that MEPAS model has the most complete exposure pathways, including the 11 exposure pathways stated in Section 2.1. MMSOILS excludes three exposure pathways: ingestion of shower water, inhalation of shower air, and inhalation of indoor air, while CalTOX does not consider pathways of shower water ingestion, soil ingestion and dermal contact of soil. All the three models follow the same principle to calculate exposure dose and human risk, which can be described by the following equation (USEPA, 1989):

$$\text{CancerRisk} = \sum_k [1 - \exp(\text{ADI}_k \times \text{CSF}_k)]$$
$$\approx \sum_k \text{ADI}_k \times \text{CSF}_k$$

where ADI_k is the average daily intake (mg kg^{-1} day^{-1}) of the contaminants by receptors through exposure route k (inhalation, ingestion, or dermal absorption), CSF_k is the cancer slope factor (kg day mg^{-1}) of exposure route k, which is derived from dose–response studies.

In practice, the linearized portion is used for small value of $\text{ADI} \times \text{CSF}$. ADI is the amount of contaminants received by human bodies via all exposure pathways, and is evaluated based on the following equation (Mckone, 1993a):

$$\text{ADI}_{ijk} = C_i \times \left[\frac{C_j}{C_i}\right] \times \left[\frac{\text{CR}_{ijk}}{\text{BW}}\right] \times \frac{\text{EF} \times \text{ED}}{\text{AT}}$$

where ADI_{ijk} (mg kg^{-1} day^{-1}) is the average daily intake from environmental media i (air, soil or groundwater), exposure media j (drinking water, food, etc.) and exposure route k (inhalation, ingestion, or dermal
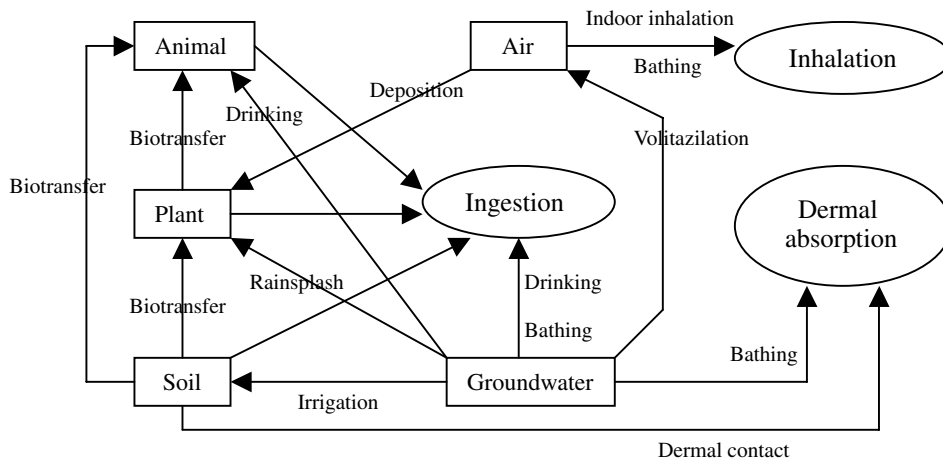

Fig. 1. The multimedia transfer processes and multiple exposure pathways associated with a contaminated-groundwater scenario.

absorption), C_i is the pollution concentration in environmental media i, C_j is the pollution concentration in exposure media j, CR_{jk} is the contact rate of exposure media j through exposure route k, EF is the exposure frequency, ED is the exposure duration, AT is the averaging time, and BW is the body weights of receptors.

The pollution concentration in exposure media, C_j, calculated from the different mathematical formulations in different multimedia models will produce different risk outcomes, and the outcomes can be the basis for choosing better models.

2.3. Uncertainty analysis and identification of important information

To reflect the uncertainty of risk estimates associated with parameters, a Monte Carlo technique is used to propagate parameter uncertainty. Each of the input parameters that are considered uncertain is treated as a random variable with known or estimated probability density function (pdf). For each of the variables, one value is selected at random with respect to the associated pdf. The individual cancer risk is then calculated by using environmental multimedia risk assessment models with any sample set of input values. The sampling and calculation are repeated 5000 times, which has been examined sufficient to avoid unacceptable variance in different realizations, to produce the pdf of the risk estimate in this case study. In this case consideration, there

are 55, 43, and 33 parameters in MEPAS, CalTOX, and MMSOILS, respectively. 33, 28, and 24 parameters, in order of the above, are treated stochastically.

Along with the Monte Carlo simulation, sensitivity analysis method is used to identify important information, whose uncertainty is a driving factor in the overall uncertainty of risk estimates for population. In this study, a rank correlation coefficient between each input parameter and the associated risk output is computed to measure the importance of each parameter to the overall uncertainty. The rank correlation coefficients are indicators of the degree of monotonic relationship between the sample values of the model prediction and those of the uncertain inputs. For this reason, rank correlation coefficients often work better to rank parameter contributions to uncertainty than other methods that are based on linear relationship only (Ma, 2002).

The following provides the procedures of screening models to reduce uncertainty due to improper model selection. Identification of important parameters with great contribution to uncertainty of risk estimates is also included to facilitate planning of information collection efforts for reducing parameter uncertainty.

(1) According to the contamination and site characteristics and exposure scenarios assumed in the practical situation, develop the conceptual site model to incorporate the relevant exposure pathways in these environmental multimedia models.

Table 1
The risk values at 95th percentile for specified pathways in the multimedia models and the differences of the modeling results

Exposure pathway	Multimedia models						The difference among models	
	MEPAS		MMSOILS		CalTOX			
	Risk	Contribution (%)	Risk	Contribution (%)	Risk	Contribution (%)	Absolute difference[a]	Relative difference[b]
Ingestion of drinking water	1.06E−06	27.35	1.09E−06	83.78	1.08E−06	48.24	3.00E−08	0.0116
Ingestion of shower water	6.08E−09	0.16	–	–	–	–	6.08E−09	0.0024
Ingestion of meat	8.76E−12	0.00	8.63E−12	0.00	7.25E−12	0.00	1.51E−12	0.0000
Ingestion of milk	6.59E−12	0.00	6.81E−12	0.00	5.65E−12	0.00	1.16E−12	0.0000
Ingestion of vegetables	1.12E−07	2.89	2.69E−08	2.07	4.97E−08	2.22	8.51E−08	0.0330
Ingestion of crop	2.21E−07	5.70	9.73E−08	7.48	9.56E−08	4.27	1.25E−07	0.0484
Ingestion of soil	2.96E−11	0.00	3.89E−14	0.00	–	–	2.96E−11	0.0000
Dermal contact of shower	4.19E−08	1.08	8.51E−08	6.55	8.23E−08	3.68	4.32E−08	0.0167
Dermal contact of soil	1.54E−09	0.03	1.76E−09	0.14	–	–	1.76E−09	0.0007
Inhalation of indoor air	9.13E−07	23.56	–	–	9.81E−08	4.38	9.13E−07	0.3539
Inhalation of shower air	1.52E−06	39.22	–	–	8.33E−07	37.21	1.52E−06	0.5891
Total risk	3.88E−06	100	1.30E−06	100	2.24E−06	100	2.58E−06	–

[a] The absolute difference is the value of difference between the estimated largest and smallest risks in each exposure pathway.
[b] The relative difference is the value of the absolute difference in each exposure pathway divided by the absolute difference of total risk.

For each multimedia model and considered exposure pathway, perform risk calculations and uncertainty analysis of parameters described above to produce probability distribution of risk estimates.

(2) Determine an appropriate percentile (e.g., 95th percentile) from the cumulative probability distributions produced in step (1). The value corresponding to the pre-determined percentile and individual exposure pathways for each model are compared and the important exposure pathways with great risk contributions are identified.

(3) The differences of the total risk values estimated by various models are used to represent model uncertainty. The relative differences of individual exposure pathway (see Table 1) among the models are also calculated to illuminate the relative magnitude of individual exposure pathway's divergence among the models. For the pathways with large divergence, the different algorithms in each model should be analyzed in order to understand the sources of model uncertainty. Further, when a model neglects an important exposure pathway, it is considered unsuitable for the case.

(4) Perform the sensitivity analysis described in Section 2.3 for the models that are not screened out in step (3) to determine the important parameters.

3. Results and discussion

3.1. The differences of the risk among the multimedia models in the specific scenario

The risk values at the 95th percentile for each exposure pathway and each environmental multimedia model, and the differences between the modeling outputs are shown in Table 1.

In this case, it is found that inhalation of shower air, ingestion of drinking water, and inhalation of indoor air are the major contributors to the total health risk (39.2%, 27.4%, and 23.6% of the total risk, respectively) in the MEPAS model. Similarly, the most important pathways identified by CalTOX model are ingestion of drinking water, inhalation of shower air, and inhalation of indoor air (48.2%, 37.2%, and 4.4% of the total risk, respectively). However, the ranks of pathways are not identical for the two models, which is due to the different mathematical formulations and will be explained later in Section 3.2.

The simulation outcome of MMSOILS model is quite different from the other two models. It is found that ingestion of drinking water (83.8% of the total risk)

is the most dominant exposure pathway. The secondly and thirdly important pathways are ingestion of crop (7.5% of the total risk) and dermal contact of shower (6.6% of the total risk). The results of Table 1 suggest that different models will indeed produce different important pathways and estimated risk outcomes, although they are assessing the same case. From the comparison of the contribution of different exposure pathways, it can be seen that the obvious diversity among the three models results from the omission of the exposure pathways of the inhalation of shower air and inhalation of indoor air in the MMSOILS model.

3.2. The analysis of model differences

From the perspective of the total risk, the risk outputs of the three models range within the same order of magnitude and the model uncertainty computed based on the comparison of the three modeling outputs is 2.58×10^{-6}. However, in order for the risk results to be useful for understanding the sources of model uncertainty and designing risk management measures, the relative importance of various exposure pathways needs to be identified.

From the aspect of comparing risk results of various pathways, the differences of the risk results for inhalation of shower air, inhalation of indoor, ingestion of crop, and ingestion of vegetables (the values of relative difference are 0.5891, 0.3539, 0.0484, and 0.0330, respectively) accounted for the large part of the differences of the total risk between models, as presented in Table 1. These exposure pathways are included in two submodels: irrigation and indoor air. The following presents the differences in calculating plant concentrations due to irrigation and indoor air concentrations of the three models. Analyzing the algorithms of these pathways in the three models could identify the sources of model uncertainty and help to choose the better multimedia models.

3.2.1. The differences in irrigation models for transferring contaminants from irrigation water to plant

Calculation of contaminants transferred from irrigation water to plant in the MEPAS model includes two processes. One is the accumulation of the contaminants in soil, and the subsequent uptake by roots from soil to edible portion of plants; the other is the direct deposition of the contaminants from irrigation water onto plant surfaces, and subsequent transfer to edible portion of plants. The accumulation of pollutants in soil over multiple years is accounted for by a soil accumulation factor (SAF). The soil accumulation factor is evaluated as the time integral of the solution to the deposition and decay differential equation, normalized to unit deposition and averaged over the deposition period

(Buck et al., 1995). The calculations are represented by Eqs. (1) and (2) in Table 2. The explanations of notation in Table 2 are attached as Appendix A. After calculating SAF, the plant contamination (C_{lvi}) resulting from irrigation deposition onto edible parts of plants and root uptake from soil is estimated through Eqs. (3) and (4) in Table 2.

Compared to MEPAS model, MMSOILS model does not consider the deposition of contaminants from irrigation water to plant surfaces. Instead, it only calculates the deposition of contaminants from irrigation water to soil and the subsequent uptake of contaminants by plant roots. The influence of recharge, soil erosion, and chemical degradation has been included in the calculation of soil concentration from irrigation water (USEPA, 1996). The leafy vegetable plant concentration (C_{vu}) is estimated from Eq. (5) in Table 2.

CalTOX model simulates chemical concentration of exposed produce sprayed with irrigation water and up-take through root directly, without calculating the soil concentration, which is the inter-medium in the transfer pathway (Mckone, 1993c). Therefore, in contrast to MEPAS and MMSOILS models, CalTOX model does not consider ingestion of soil and dermal contact of soil, although both pathways are explicitly addressed in Cal-TOX model. The chemical concentration of exposed produce is calculated by Eq. (6) in Table 2.

3.2.2. The differences in indoor air models

Two submodels are included in the MEPAS model for calculating the risk from indoor inhalation of volatile pollutants: the MEPAS shower inhalation model and the USEPA Andelman indoor inhalation model. Although the concentration in shower air can be estimated by using Henry's law constant, the maximum allowable Henry's law constant is restricted to $2.4 \times 10^{-3} \, \text{m}^3 \, \text{atm} \, \text{g}^{-1} \, \text{mol}^{-1}$ due to limitation of mass balance. Because both of Henry's law constant of TCE

Table 2
The differences of the mathematical formulations in MEPAS, MMSOILS, and CalTOX models

Submodel	Model	Mathematical equations
Irrigation model from irrigation water to plant	MEPAS	(1) $\dfrac{dC_{awi}}{dt} = UD_{wi} - C_{awi}\lambda_{di}$, and (2) $SAF_i = \dfrac{\int_0^{ED_{KK}365.25} C_{awi}\,dt}{UD_{wi}ED_{KK}365.25}$
		(3) $C_{lvi} = (CWD_{lvi} + CWR_{lvi})e^{-\lambda_{gi}TH_{lv}}$ $= \left[DP_{wi}TV_{lv}r_{lv}\dfrac{(1-e^{-\lambda_{ei}TC_{lv}})}{\lambda_{ei}Y_{lv}} + \dfrac{FI_{lv}SAF_iB_{vi}DP_{wi}}{P} \right]e^{-\lambda_{gi}TH_{lv}}$ and (4) $DP_{wi} = \dfrac{C_{iri}IR}{30}$
	MMSOILS	(5) $C_{vu} = B_vf_{dw}C_{as} = B_vf_{dw}\dfrac{C_{irr}D_{irr}XF6}{\rho_bD_m\lambda_s}$
	CalTOX	(6) $C_{fvg}^{abgw} = C_q TF(q \to ep) = C_qf_q^w(K_{ps}^{rain} + K_{ps})K_Df_{ir}$
Indoor air model	MEPAS	(7) Showering $C_{sai} = 10^3 C_{dwi}TF_i e^{-\lambda_{gi}TH_{dw}}\left(\dfrac{H_i}{RT}\right)$
		(8) Indoor $C_{iai} = C_{dwi}TF_i e^{(-\lambda_{gi}TH_{dw})}K_c$
	MMSOILS	No consideration
	CalTOX	(9) Showering $C_{ba} = C_q TF(q \to bathair) = C_qf_q^w\dfrac{W_{bath}}{VR_{bath}}\phi_x(bath)$ $= C_qf_q^w\dfrac{W_{bath}}{VR_{bath}}\dfrac{1.8 \times 10^5}{\left[\dfrac{2.5}{D_l^{2/3}} + \dfrac{RT}{H \cdot D_a^{2/3}}\right]}$
		(10) Indoor $C_{ia} = C_q TF(q \to inair) = C_qf_q^w\dfrac{W_{house}}{VR_{house}}\phi_x(house)$ $= C_qf_q^w\dfrac{W_{house}}{VR_{house}}\dfrac{2.1 \times 10^5}{\left[\dfrac{2.5}{D_l^{2/3}} + \dfrac{RT}{H \cdot D_a^{2/3}}\right]}$

and PCE in this case study are greater than $2.4 \times 10^{-3} \, m^3 \, atm \, g^{-1} \, mol^{-1}$, this restricted value has been adopted. The shower air concentration is calculated by Eq. (7) in Table 2 (Lyman et al., 1982). For calculating the concentration in indoor air from domestic water, the following conditions must be met: Henry's law constant is greater than $10^{-5} \, m^3 \, atm \, g^{-1} \, mol^{-1}$ and molecular weight is smaller than $200 \, g^{-1} \, mol$. The chemical concentration in the air in the home is evaluated as Eq. (8) in Table 2 (Andelman, 1990).

Because the atmospheric pathway in the MMSOILS model only considers the release of contaminants from the site in the form of vapors and fugitive dust emissions from wind erosion and mechanical disturbances, both of the important pathways, inhalation of shower air and inhalation of indoor air due to contaminants vaporizing from domestic water, are not evaluated. This is the essential reason that MMSOILS model deviates from the other two models in the case.

Although the calculation of indoor inhalation in the CalTOX model includes the same pathways as MEPAS, their mathematical formulations of the pathways are not the same. The formulations in the CalTOX are taken from Mckone and Bogen (1992) and are represented by Eqs. (9) and (10) in Table 2.

From the proceeding description of the formulations, the transfer pathways and mathematical equations of the three models related to contaminants from domestic

water to indoor air and bathroom air are all different. The MMSOILS model even completely omits the indoor air model. It is apparent that the modeling divergence of pathways related to irrigation and indoor inhalation leads to the differences of the estimated risk results.

Therefore, before performing a risk assessment, the model uncertainty should be evaluated by first focusing on the relation between conceptual model and multimedia models, and on the significance of the neglected pathways and differences of mathematical formulations between multimedia models.

3.3. The selection of the better models through model comparison and reduction of model uncertainty

The relative importance of neglected pathways in the three models can be obtained from the value of relative difference in Table 1. While the MEPAS model has incorporated the 11 exposure pathways in the conceptual model, CalTOX model omits three pathways, including ingestion of shower water, ingestion of soil, and dermal contact of soil, which are shown not relatively significant, because their values of relative difference are 0.0024, 0.0000, and 0.0007, respectively. MMSOILS model eliminates the pathways of ingestion of shower water, inhalation of indoor air, and inhalation of shower air. Based on the values of relative difference shown in Table 1 (0.0024, 0.3539, and 0.5891,

Table 3
The risk values for specified pathways and the differences of the modeling results without Monte Carlo simulation

| Exposure pathway | Multimedia models | | | | | | The difference among models | |
| | MEPAS | | MMSOILS | | CalTOX | | | |
	Risk	Contribution (%)	Risk	Contribution (%)	Risk	Contribution (%)	Absolute difference[a]	Relative difference[b]
Ingestion of drinking water	3.96E−07	28.68	3.95E−07	91.05	4.01E−07	50.33	4.00E−09	0.0042
Ingestion of shower water	1.99E−09	0.14	–	–	–	–	1.99E−09	0.0021
Ingestion of meat	2.39E−12	0.00	2.40E−12	0.00	2.60E−12	0.00	2.10E−13	0.0000
Ingestion of milk	1.91E−12	0.00	1.93E−12	0.00	2.08E−12	0.00	1.70E−13	0.0000
Ingestion of vegetables	1.38E−08	1.00	2.51E−09	0.58	1.52E−08	1.91	1.27E−08	0.0134
Ingestion of crop	2.43E−08	1.76	9.83E−09	2.27	2.96E−08	3.72	1.98E−08	0.0209
Ingestion of soil	3.61E−12	0.00	3.92E−15	0.00	–	–	3.61E−12	0.0000
Dermal contact of shower	1.53E−08	1.10	2.63E−08	6.06	2.62E−08	3.29	1.10E−08	0.0116
Dermal contact of soil	1.80E−10	0.01	1.95E−10	0.04	–	–	1.95E−10	0.0002
Inhalation of indoor air	3.93E−07	28.45	–	–	4.07E−08	5.11	3.93E−07	0.4145
Inhalation of shower air	5.37E−07	38.85	–	–	2.84E−07	35.65	5.37E−07	0.5677
Total risk	1.38E−06	100	4.34E−07	100	7.97E−07	100	9.46E−07	–

[a] The absolute difference is the value of difference between the estimated largest and smallest risks in each exposure pathway.
[b] The relative difference is the value of the absolute difference in each exposure pathway divided by the absolute difference of total risk.

respectively), the pathways of inhalation of indoor air and shower air are very important. Therefore, although both CalTOX and MMSOILS exclude three pathways, the omissions in CalTOX model does not significantly impact the total risk in this case.

The preceding comparison of modeling results reveals that MMSOILS is improper for the case because it neglects the exposure pathways that are deemed important in the other two models. We can further check that the total risk (1.30×10^{-6}) in the MMSOILS model is similar to the total risk (1.44×10^{-6}) in the MEPAS model, if the risk of ingestion of shower water, inhalation of indoor air, and inhalation of shower air is subtracted from MEPAS modeling outputs. Likewise, when the risk of inhalation of indoor air and inhalation of shower air is subtracted from CalTOX modeling outputs, the total risk (1.31×10^{-6}) in CalTOX model is almost the same as the total risk in the MMSOILS model.

Therefore, MEPAS and CalTOX model are more suitable than MMSOILS in this case. In other words, eliminating unsuitable MMSOILS model can effectively decrease model uncertainty. In this case, the model uncertainty decreases from 2.58×10^{-6} to 1.64×10^{-6}, equaling 36.4% reduction of model uncertainty.

3.4. The importance of Monte Carlo simulation

In contrast to the risk values at 95th percentile calculated in Table 1, Table 3 presents the results of risk values without using Monte Carlo technique to propagate parameter uncertainty. The model uncertainty is 9.46×10^{-7} and 38.4% of the model uncertainty can be reduced. Although the values in Table 3 are all smaller than those in Table 1 and the ranks of the relative difference in Table 3 are the same as those in Table 1, the risk values of four foodchain pathways in CalTOX model become greater than those in the other two models in Table 3, which is due to the smaller parameter uncertainty of foodchain pathways in CalTOX model. Although the selection of better models is not changed in the case, the mentioned outcome obviously reveals that the parameter uncertainty may interfere with selection of better models in other cases.

3.5. The determination of the major parameters from the better models

The results from the sensitivity analysis for identifying critical parameters are described in Table 4. It is found that the numbers of important parameters (contribution to variance of total risk greater than 0.5%) in MEPAS, MMSOILS, and CalTOX model are 8, 4, 6 respectively. The results show that TCE concentration in groundwater, PCE concentration in groundwater, ingestion rate of drinking water, and body weight are the four important parameters that are identified by

Table 4
The major parameters from the sensitivity analysis in the three multimedia models

Parameter	Definition	Contribution to variance of total risk			Distribution	Reference
		MEPAS (%)	MMSOILS (%)	CalTOX (%)		
C_{wt}	TCE concentration in groundwater (mg l⁻¹)	64.9	17.9	39.1	Normal (mean = 7.41E−4, SD = 9.31E−4)	Site survey
TEs	Exposure time during shower (h)	11.4	–	3.1	Lognormal (mean = 0.167, GSD = 1.7)	Droppo et al. (1989)
C_{wp}	PCE concentration in groundwater (mg l⁻¹)	8.9	29.0	26.1	Normal (mean = 9.50E−5, SD = 2.85E−4)	Site survey
BW	Body weight (kg)	6.6	5.0	4.0	Normal (mean = 59.9, SD = 10.6)	Kao et al. (1997)
U_{dw}	Ingestion rate of drinking water (l d⁻¹)	3.9	47.7	25.3	Lognormal (mean = 2.0, GSD = 1.7)	Droppo et al. (1989)
U_{si}	Inhalation rate of shower (m³ d⁻¹)	1.0	–	–	Lognormal (mean = 2.0, GSD = 1.2)	Droppo et al. (1989)
K_{pp}	Skin absorption permeability constant for PCE (cm h⁻¹)	0.7	–	–	Lognormal (mean = 0.0488, GSD = 2.0)	CalEPA (1993)
K_c	Indoor volatilization factor (l m⁻³)	0.5	–	–	Lognormal (mean = 0.5, GSD = 1.2)	Droppo et al. (1989)
foc_s	Organic carbon fraction in surface soil (dimensionless)	–	–	0.5	Lognormal (mean = 0.005, GSD = 1.2)	Site survey

the three models simultaneously, but with different ranks. However, exposure time during shower is an important parameter that is only recognized by MEPAS and CalTOX models. This difference results from the neglect of the important pathway of inhalation of shower air in the MMSOILS model. Therefore, the critical parameters identified by the more suitable models chosen from model comparison are more useful in designing uncertainty reduction measure.

4. Conclusion

In the research, three environmental multimedia models combined with conceptual model development and Monte Carlo simulation are applied to a contaminated-groundwater site to demonstrate the processes of model comparison in screening unsuitable models. In the light of significant uncertainty due to model selection, developing such a screening procedure would facilitate uncertainty reduction. Based on the proper model, the identification of important parameters and pathways and design of risk management measures would be better-formed. In this case study, it is found that different consideration of exposure pathways and related mathematical formulations indeed produces different modeling results and that the comparison of the major exposure pathways among models would reveal the suitability of the models. It has also been demonstrated that the model uncertainty is decreased. Furthermore, the sensitivity analysis along with the use of more proper models could facilitate reduction of parameter uncertainty by identifying a better set of important parameters, which may not be considered in an improper model.

Although the results of this particular case study cannot be generalized, the procedure provided in this study can be applied to different cases. The procedure first involves the development of conceptual model for a risk problem. The compatibility of potential candidate models with the conceptual model and comparison of various modeling performance regarding individual exposure pathways are performed by combining Monte Carlo simulation with the models. Then the source of model uncertainty can be identified by checking the model algorithms for the pathways with large contribution to the divergence among models. Finally, the model that neglects the pathways deemed important by most candidate models is considered unsuitable.

Traditionally, executing a model with Monte Carlo simulation can reveal the quantity of parameter uncertainty, but the model uncertainty would not be properly addressed. This procedure of screening models provides a means of reducing model uncertainty. It should also be noted that the parameter uncertainty associated with various models may affect the selection of better models

and therefore Monte Carlo analysis needs be used explicitly. The procedure provided in this study is believed to facilitate the reduction of model and parameter uncertainties.

Appendix A. Notations

B_v, B_{vi} soil-to-plant concentration factor, kg soil kg^{-1} dry plant or kg dry soil kg^{-1} wet weight plant

C_{fvg}^{abgw} pollutant concentration in above ground vegetables due to irrigation water, $mg\,kg^{-1}$

C_{as} soil concentration in agricultural field, $mg\,kg^{-1}$

C_{awi} soil concentration from irrigation water deposition as a function of time, $mg\,m^{-2}$

C_{dwi} pollutant concentration in domestic water, $mg\,l^{-1}$

C_{iai}, C_{ia} pollutant concentration of indoor air outside the bathroom, $mg\,m^{-3}$

C_{iri}, C_{irr}, C_q pollutant concentration in irrigation water, $mg\,l^{-1}$

C_{lvi} leafy vegetable plant concentration at time of consumption, $mg\,kg^{-1}$

C_{sai} pollutant concentration in shower air, $mg\,m^{-3}$

CWD_{lvi} pollutant concentration in leafy vegetables from water deposition onto plants, $mg\,kg^{-1}$

CWR_{lvi}, C_{vu} pollutant concentration in leafy vegetables from root uptake, $mg\,kg^{-1}$

D_a contaminant diffusion coefficient in air, $m^2\,s^{-1}$

D_{irr} application rate of irrigation, $m\,yr^{-1}$

D_l contaminant diffusion coefficient in water, $m^2\,s^{-1}$

D_m mixing depth of soil in agricultural field, m

DP_{wi} rate of deposition in irrigation water to cropland soil and plants, $mg\,m^{-2}\,d^{-1}$

ED_k exposure duration for pathway k, yr

f_{dw} fraction of wet plant remaining as dry material, dimensionless

FI_{lv} fraction of year that irrigation occurs for leafy vegetables, dimensionless

f_{ir} fraction of the pollutant concentration in irrigation water retained in soil water, unitless

f_q^w fraction of water irrigation needs provided by ground water (assumption for 1.0), unitless

H, H_i Henry's law constant, $Pa\,l\,mol^{-1}$ or $m^3\,atm\,g^{-1}\,mol^{-1}$

IR irrigation water application rate during irrigation periods, $l\,m^{-2}\,month^{-1}$

K_c Andelman volatilization factor for chemical pollutants, $l\,m^{-3}$

K_D soil/soil–water partition coefficient, kg water kg^{-1} soil solids

K_{ps} plant–soil partition coefficient from soil to plant parts due to uptake through roots, $kg\,kg^{-1}$

K_{ps}^{rain} plant–soil partition coefficient for surface soil due to rainsplash, kg soil kg^{-1} plant fresh mass

P area soil density of farmland, kg m^{-2}

R gas law constant, m^3 atm g^{-1} mol^{-1} K^{-1} or Pa l mol^{-1} K^{-1}

r_{lv} fraction of deposition retained on plant surfaces, dimensionless

SAF_i soil accumulation factor for an exposure duration of ED$_k$ years, d

T average absolute water temperature in the shower or house, K

TC_{lv} duration of the growing period for leafy vegetables, d

TF_i water treatment purification factor, dimensionless

TF(q → ep)   radio of pollutant conc. of exposed produce to the pollutant conc. in groundwater, unitless

TF(q → inair)   radio of pollutant conc. of indoor air to the pollutant conc. in groundwater, unitless

TH_{dw} holdup time in transfer from well to the consumer, d

TH_{lv} holdup time between harvest of leafy vegetables and consumption by humans, d

TV_{lv} translocation factor from plant surfaces to edible parts for leafy vegetables, dimensionless

UD_{wi} unit deposition rate to soil, mg m^{-2} d^{-1}

W_{bath} water use rate for showering, l h^{-1}

W_{house} water use rate for all household activities, l h^{-1}

VR_{bath} average bathroom ventilation rate, m^3 h^{-1}

VR_{house} average house ventilation rate, m^3 h^{-1}

Y_{lv} yield of leafy vegetables, kg m^{-2}

λ_{di} environmental degradation and decay constant in surface soil, d^{-1}

λ_{ei} effective weathering and decay constant, d^{-1}

λ_{gi} decay rate constant in closed water system, d^{-1}

ρ_b bulk density, g cm^{-3}

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