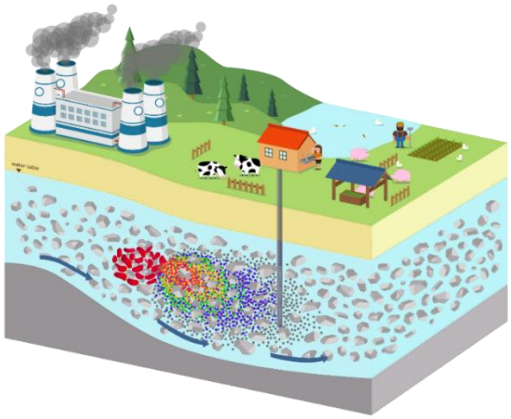
MUSt User's Manual

MUltiSpecies Transport analytical model



Jui-Sheng Chen
National Central University
Graduate Institute of Applied Geology

Ching-Ping Liang
Fooyin University
Department of Nursing

Heejun Suk
Korea Institute of Geoscience and Mineral Resources

Zhong-Yi Liao
National Central University
Graduate Institute of Applied Geology

Chen-Wuing Liu
National Taiwan University
Department of Bioenvironmental Systems Engineering

Cheng-Wen Chen
National Central University
Graduate Institute of Applied Geology

Contents

Contents 2

1. Software installation 4

2. Introduction 9

2.1 Development of the model 9

2.2 Features of the MUSt 11

3. User interface input 13

4. User interface output 20

5. References 30

Appendix A. Introduction of the analytical model 31

A.1 Governing equations for solute transport 31

A.2 Initial conditions and boundary conditions 33

A.3 Health risk assessment module 36

Appendix B. Case study 38

Case 1. Single species transport: Benzene, Toluene, Ethylbenzene, Xylene 39

Case 2. Single species transport: Radionuclide U-234 41

Case 3. Single species transport: Effects of the dispersion coefficient 43

Case 4. Multispecies transport: Three nitrogen transformation products 45

Case 5. Multispecies transport: Three radionuclides **47**

Case 6. Multispecies transport: Four radionuclides **50**

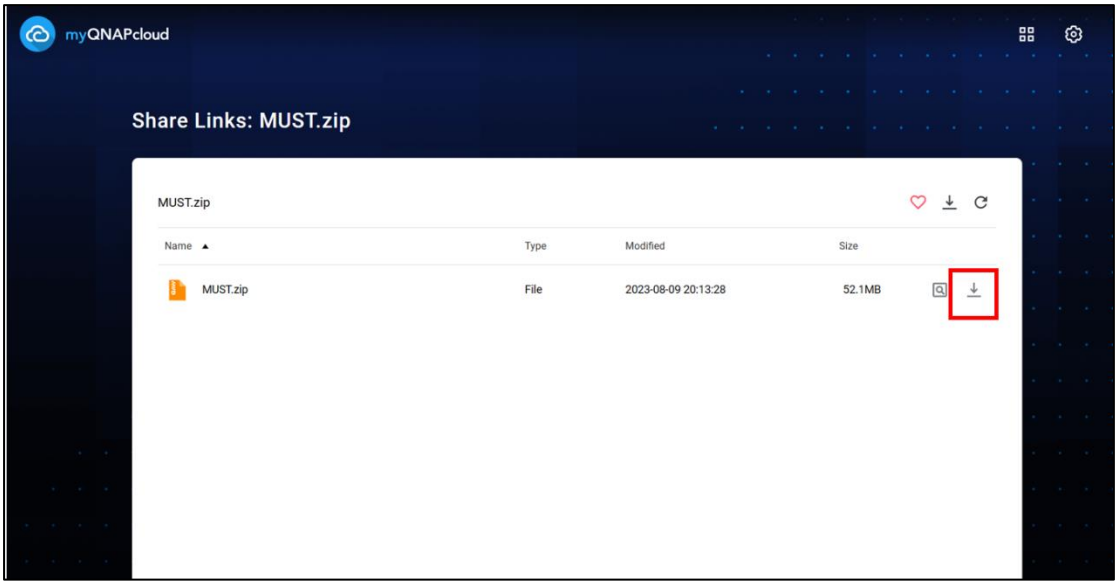
Case 7. Multispecies transport: Five chlorinated solvents **52**

Case 8. Multispecies transport: Source geometry and numbers **59**

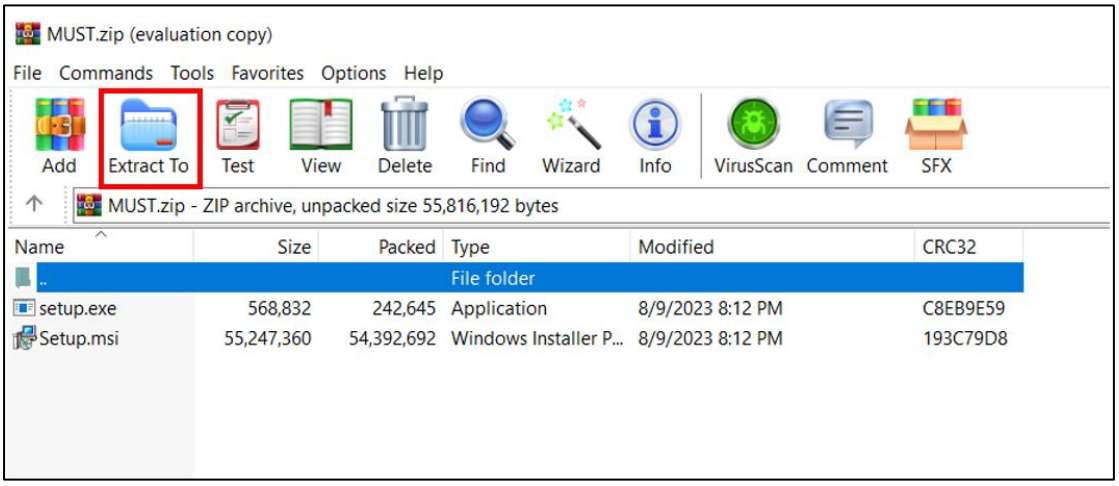
Case 9. Multispecies transport: Application of time-dependent source functions **62**

1. Software Installation

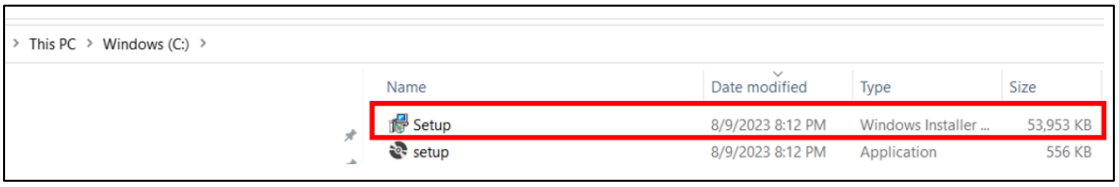
To begin, please download the software file.

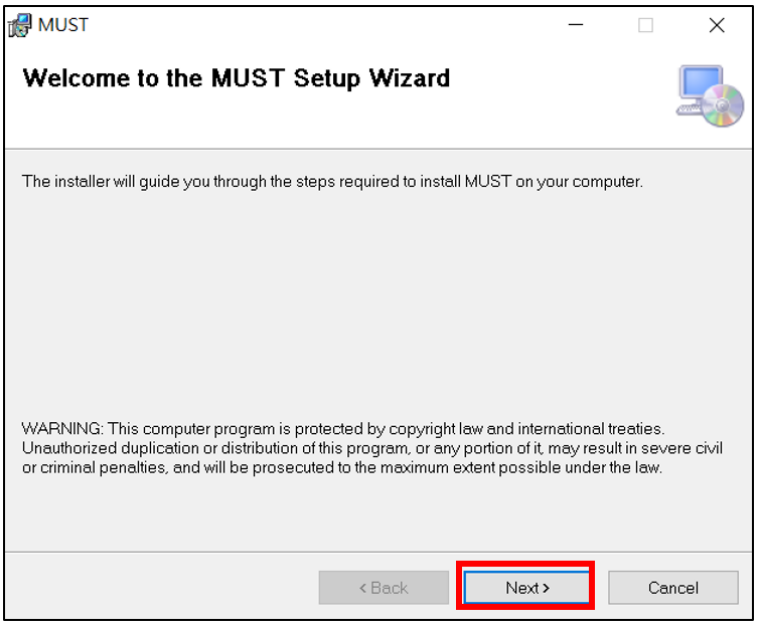


Next, extract the downloaded file to your preferred location (it is not recommended that the file be extracted to the desktop).

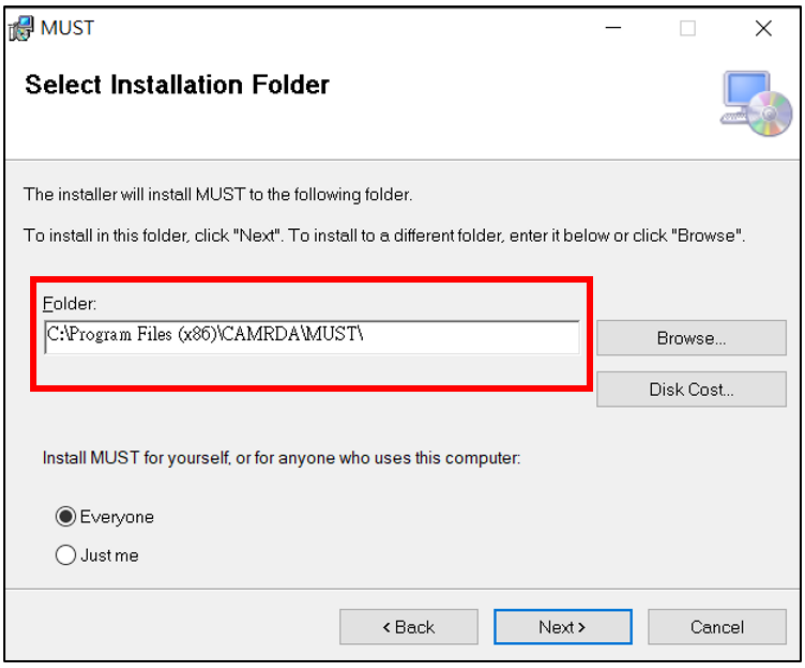


After extracting the files, click on “Setup” to proceed with the installation.

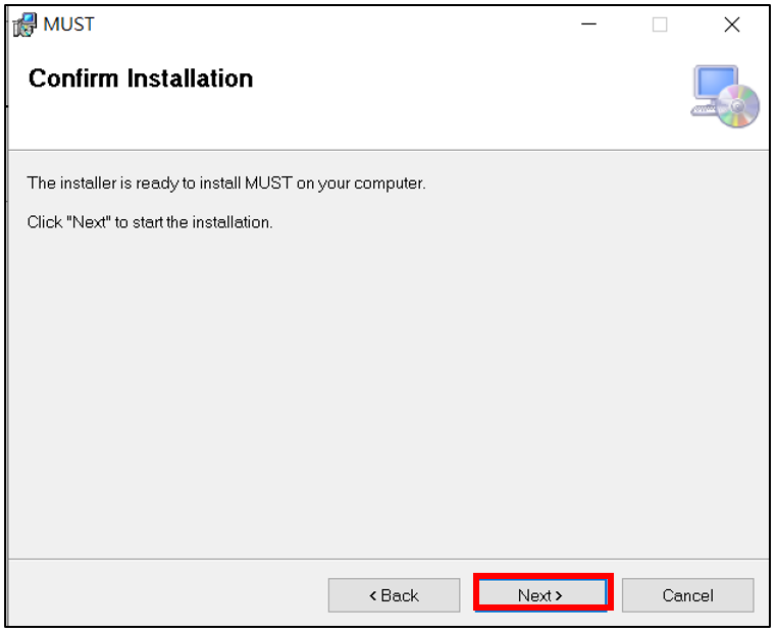




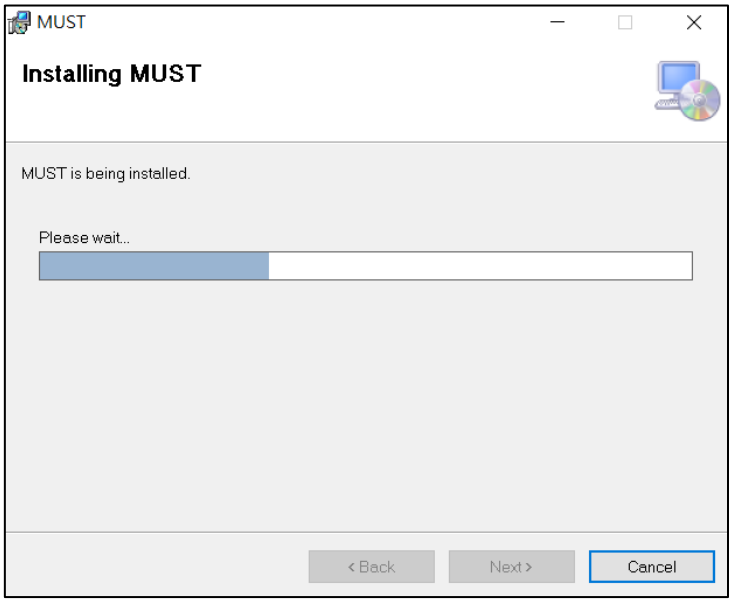
The MUSt folder should be installed in the “C:\Program Files (x86)” directory in your Windows system. If the default location is different, first click “Browse,” select the correct location, then, click the “Next” button to continue with the next step.



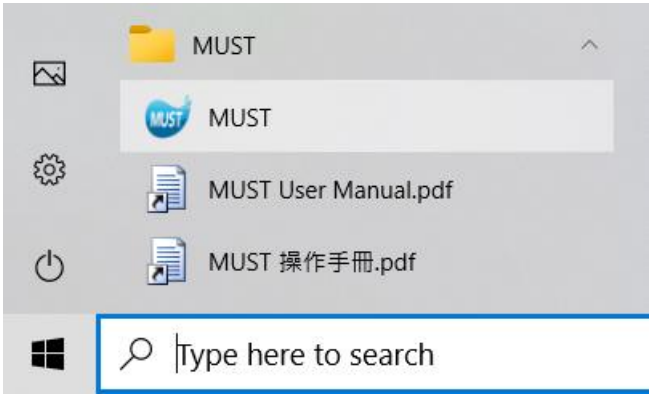
Click “Next” to proceed with the installation of the MUSt software on your computer.



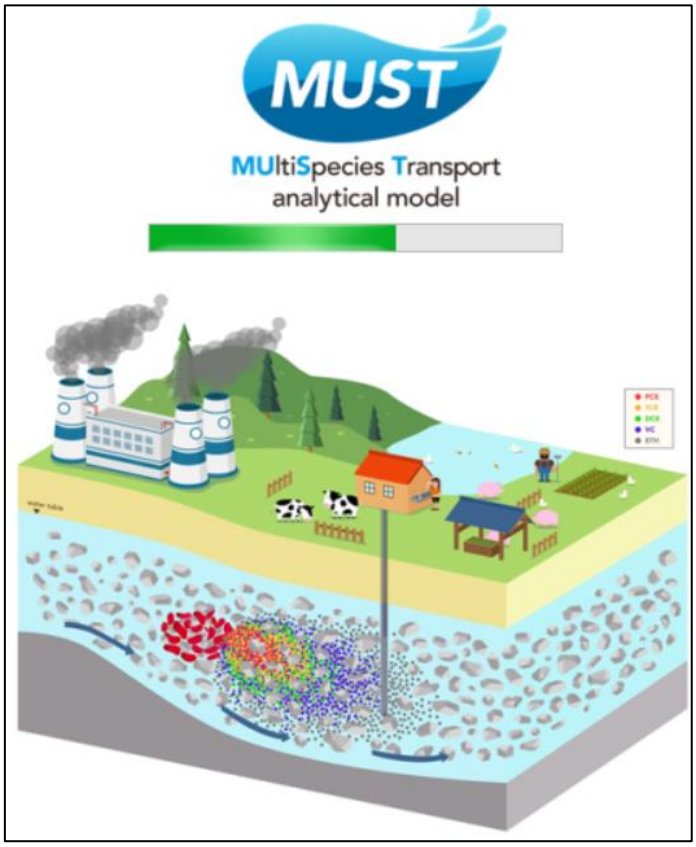
During the installation process, a progress bar will be visible, showing the progress until the installation is successfully completed.



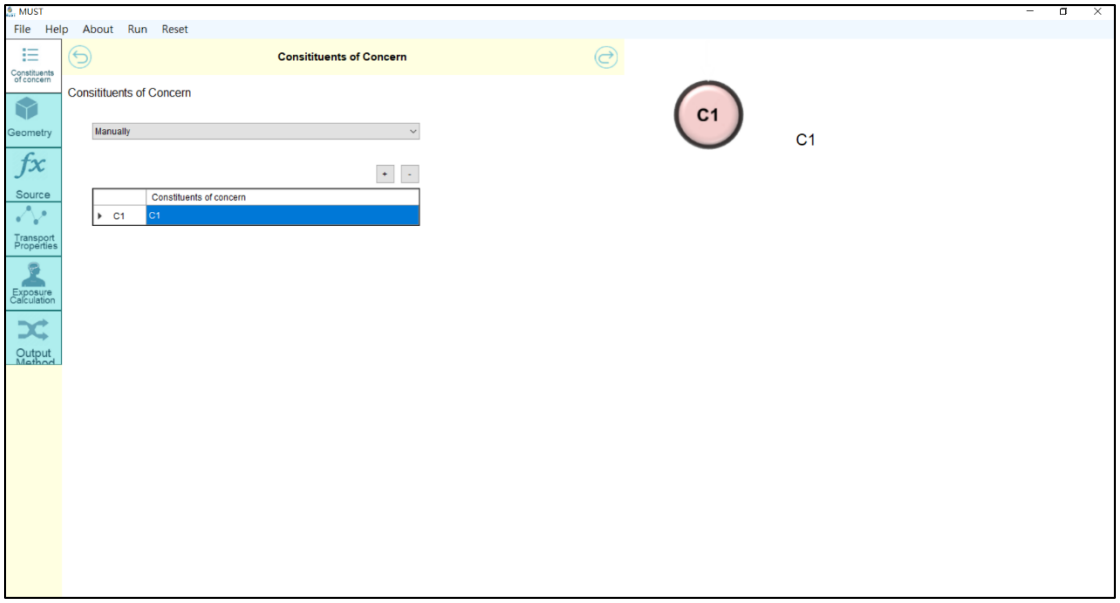
Once the installation is finished, you will find the program named “MUST” in the Start menu. To open the software, simply locate “MUST.exe” and click on it.



Upon first launch, you will be prompted to provide the license information.



Following input of the license information, you can open the software and begin using it.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2. Introduction

2.1 Development of the model

In recent years, groundwater pollution has become a very important environmental issue. The existence of such pollutants in the environment for long periods of time increase the risk of irreparable harm to the environment and human health. It is very important to take steps to ensure that contaminant concentrations conform to the regulatory standards. In addition, since so many groundwater contaminants are carcinogenic, it is also important to quantify the risk to human health when identifying standards for the assessment of contaminated sites.

Analytical mathematical models based on physical mechanisms are quite effective tools for understanding the fate and transport of contaminants in groundwater. In addition, analytical models are also very useful for parameter estimation, pollution control, pollution remediation, and human health risk assessment for contaminated sites. In the past, although there have been some developments of software formulated using an analytical model such as AT123D (Yeh, 1981), BIOSCREEN (Newell et al., 1996), HYDROSCAPE (Funk et al., 2017), the software has all been based on the simple advective-dispersive transport equation (ADE) shown in Eq. (1), which can only simulate the transport of a single contaminant.

$$R\frac{\partial C}{\partial t} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} + D_z \frac{\partial^2 C}{\partial z^2} - v\frac{\partial C}{\partial x} - kC \tag{1}$$

Here, C is the concentration of the solute in dissolved phase; x, y, z are the spatial coordinates; t is the time; R is the retardation factor of the contaminants; D_x is the longitudinal dispersion coefficient in the x direction; D_y and D_z are the transverse and vertical dispersion coefficients, respectively; v is the average groundwater seepage velocity; k is the first-order decay reaction rate constant of contaminants.

However, the degradation or decay of most common groundwater contaminants including chlorinated organic solvents, radionuclides, nitrogen conversion products, etc., are often involve complex, producing multiple different toxic products in the decay

chain. Table 1 shows the common contaminant decay chain example.

Table 1. Examples of first-order decay/degradation chains of radionuclides, chlorinated solvents, and nitrogen conversion products.

Contaminants	Decay chain
Chlorinated solvent	Tetrachloroethylene (PCE) → Trichloroethylene (TCE) → Dichloroethylene (DCE) → Vinyl chloride (VC) → Ethylene (ETH)
Radionuclides	Plutonium-238 (Pu-238) → Uranium-234 (U-234) → Thorium-230 (Tu-230) → Radium-226 (Ra-226)
Nitrogen	Ammonium (NH_4^+) → Nitrite (NO_2^-) → Nitrate (NO_3^-)

A few software packages based on analytical models have been developed that can simultaneously describe the transport of multiple contaminants, for example, BIOCHLOR (Aziz et al., 2000), REMChlor (Falta, 2007), DECAY (Wang and Neville, 2019), etc., which couple several partial differential equations as shown in Eq. (2), where $g_{i-1\to i}$ is the mass conversion factor, which considers the mass conversion from the parent species to daughter species.

$$R_i \frac{\partial C_i}{\partial t} = D_x \frac{\partial^2 C_i}{\partial x^2} + D_y \frac{\partial^2 C_i}{\partial y^2} + D_z \frac{\partial^2 C_i}{\partial z^2} - v \frac{\partial C_i}{\partial x} - k_i C_i + g_{i-1\to i} k_{i-1} C_{i-1}, \quad (2)$$
$$i = 1, 2, \ldots N; \qquad y_{0\to 1} = 0$$

Among these models, DECAY can simulate only relatively simple one-dimensional transport. Although the BIOCHLOR (Aziz et al., 2000) and REMChlor (Falta, 2007) models can simulate three-dimensional scenarios closer to real-world situations, both can only consider the same retardation factor when describing the sorption of different contaminants.

Recently, Liao et al. (2021) developed a new three-dimensional transport analytical model which not only improves the limitation on the use of the same retardation factor for all contaminants but also has a very high computational efficiency. A schematic diagram of their three-dimensional model is shown in Figure 1, where the rectangular patch area is the source zone. They derived analytical solutions written in

the Fortran programming language.

The MUSt software developed by our team is based on this new model and also features a user-friendly graphical user interface, which allows users to simulate the transport of a single or multiple contaminants at the same time, such as for predicting the transport distribution of chlorinated organic solvents and radionuclides in the subsurface environment. In addition, the software also includes a health risk calculation module, which can be used to calculate the carcinogenic and non-carcinogenic risks of the various contaminants to the human body. This MUSt software will be a very effective tool for the practical applications, health risk assessment and contaminated site remediation.

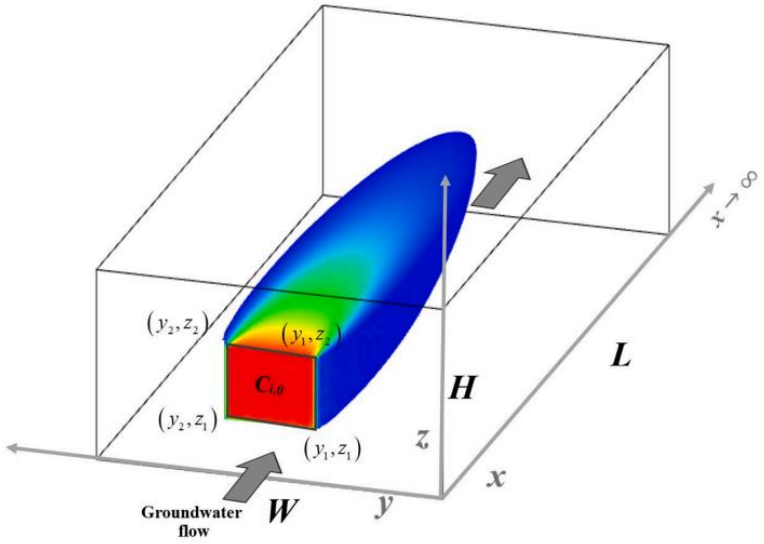


Fig. 1. Schematic view of the three-dimensional migration of a multispecies plume in a groundwater system produced by the MUSt software (from Liao et al. (2021)).

2.2. Features of the MUSt

1. Three different types of source functions can be considered, a constant concentration, exponential decay, and piecewise constant concentrations that can be input according to the field data at different times.

2. Multiple source areas can be established. Different concentrations for different

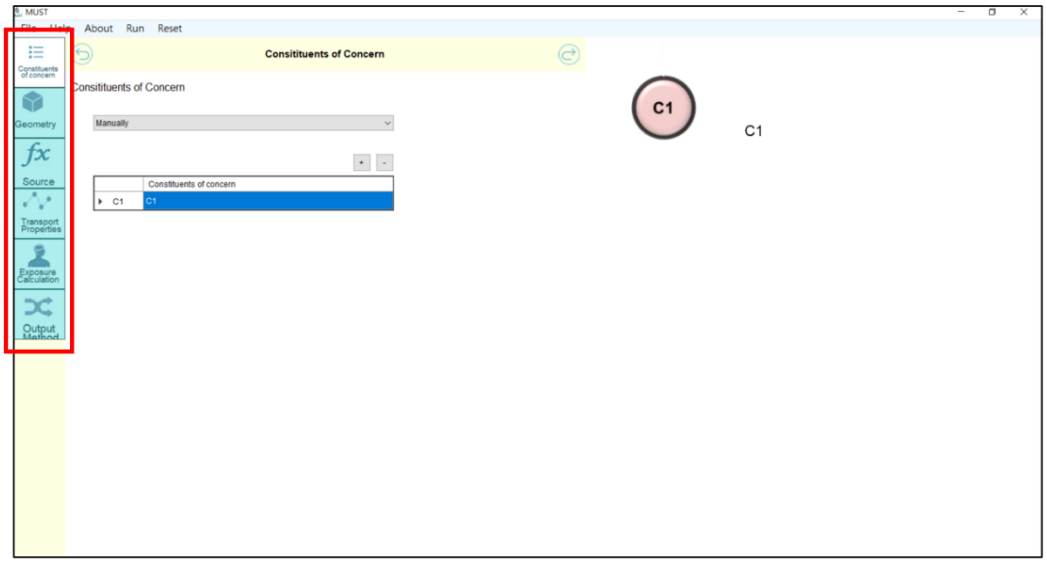
contaminants can be input in different areas; this is more practical for describing the geometric shape and concentration distribution of different contaminant sources.

3. The health risk assessment can be calculated, showing the high, medium and low risk areas near the site based on the defined standard values.

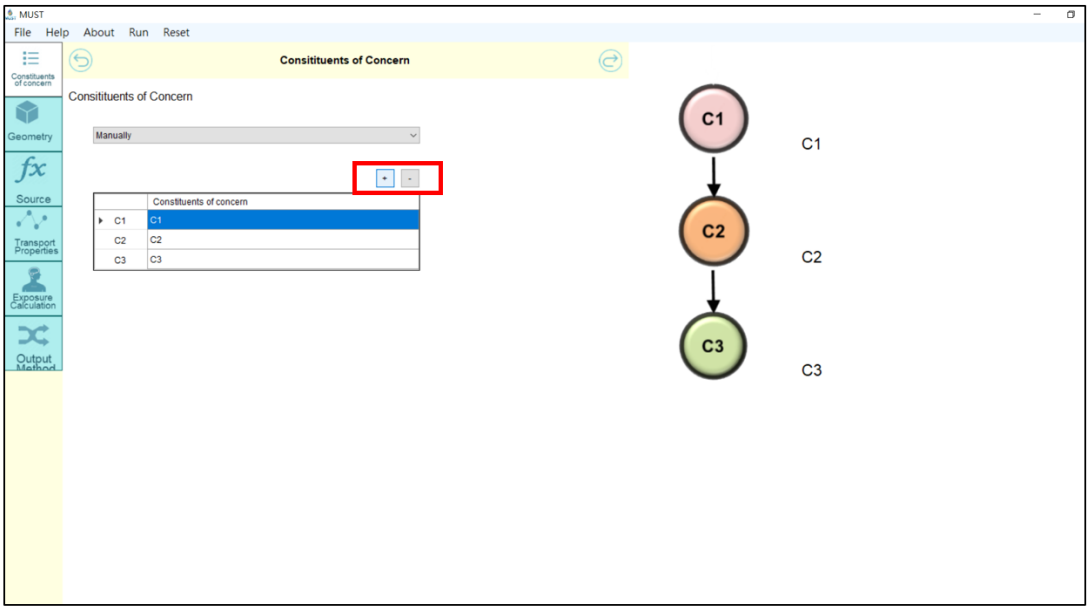
4. The software offers a variety of methods for visualization of the output results, including the breakthrough curve of the observation point, the concentration profile along different directions, the distribution of plume migration at different depths, an animation of plume migration at different times, and tables listing the concentration values.

3. User interface input

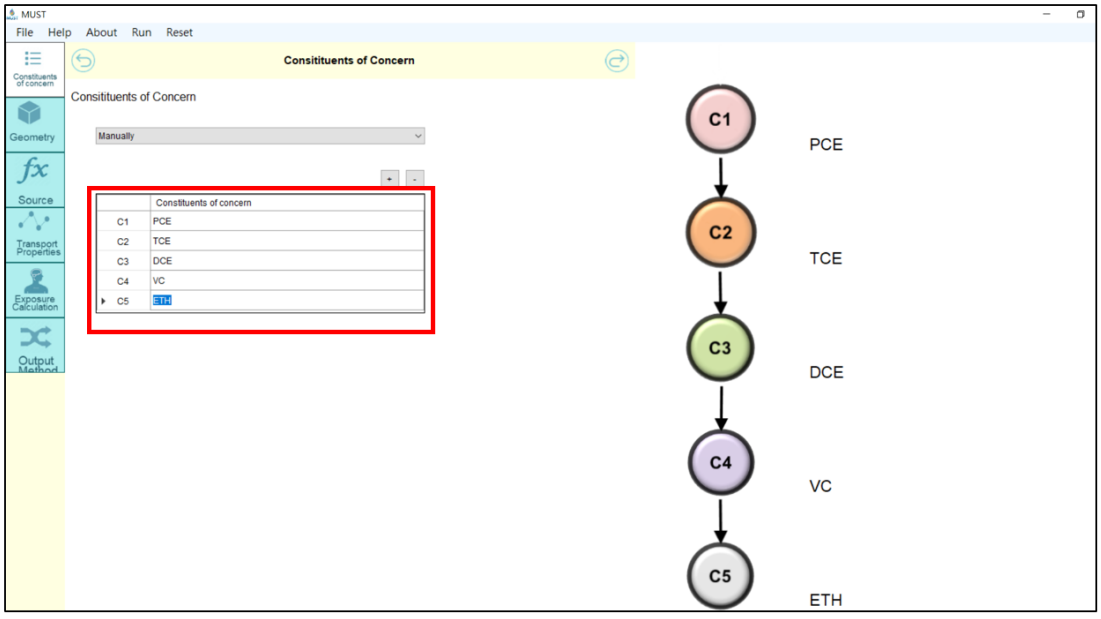
This chapter provides a step-by-step guide on how to complete parameter input through the software interface. When the MUSt software is launched, the main tab depicted below will appear. Note the six square buttons on the left-hand side, labelled based on the parameter properties, to facilitate user input and case construction. These buttons are organized into six sequential steps, from top to bottom. The first five steps cover parameter input relevant to the model simulation; the sixth step specifies the output visualization options. The tab or screen to then right of the square buttons details the currently selected step. The left half of the screen is dedicated to parameter input and option selection, while the right half serves as the interactive area. This area is adjusted based on the user's input and provides relevant supplementary descriptions as needed.



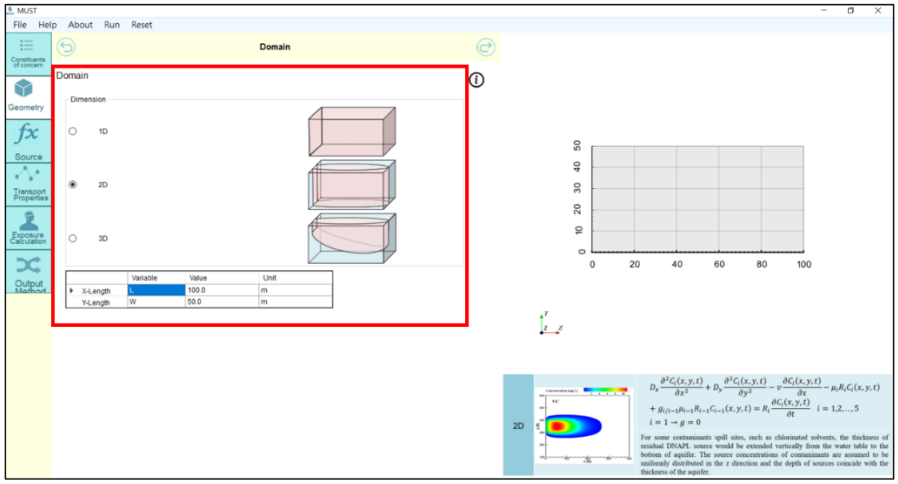
In this example, we will use a two-dimensional scenario to illustrate the transport of chlorinated solvents. The initial step involves defining the contaminants of concern. Depending on the specific case, the user has the flexibility to adjust the number of contaminants in the reaction decay chain, as well as assign names to these contaminants as needed.



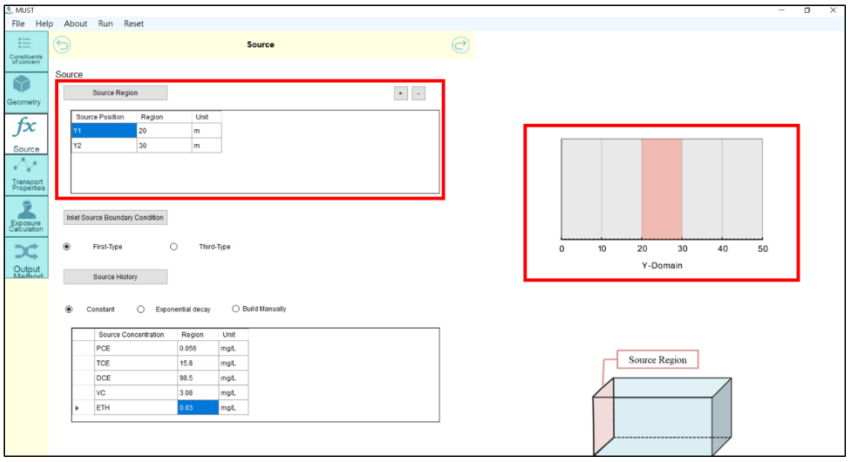
The contaminants used in this case are the chlorinated solvent, tetrachlorethylene, and its degradation products. There are five species $PCE \rightarrow TCE \rightarrow DCE \rightarrow VC \rightarrow ETH$ in total, so the "+" button is pressed four times to increase the number of contaminants to five.



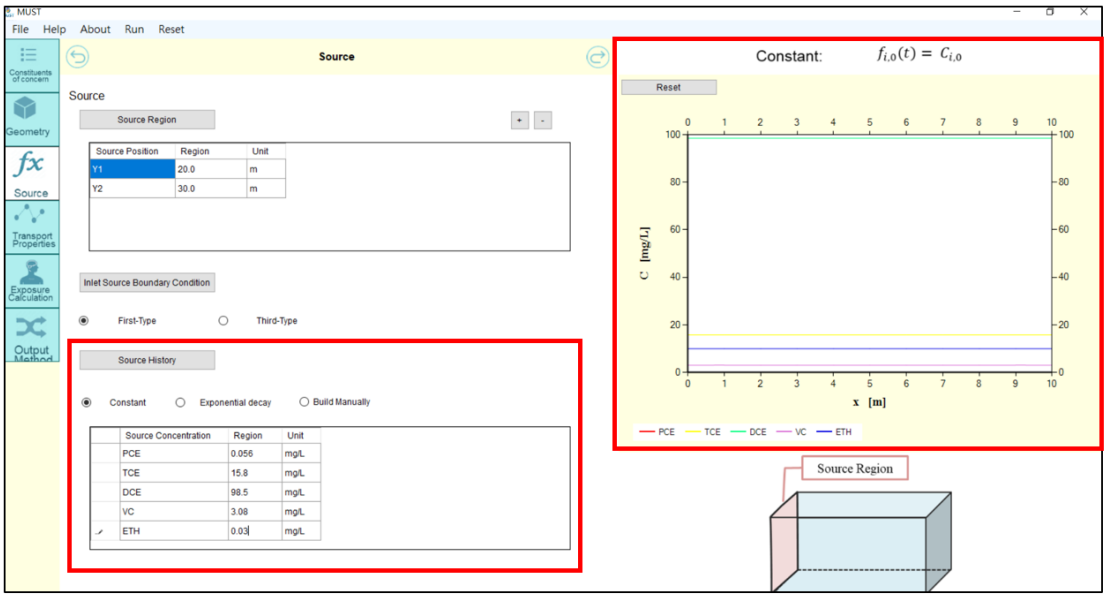
The second step involves setting up the aquifer scenario. In the options, the user can select either the simpler one-dimensional simulation or opt for the more complex two- and three-dimensional scenario simulations. First, define the dimensions of the aquifer geometry, specifying the length, width, and height. In this instance, since this is a two-dimensional scenario, click on “2D.” The interactive screen on the right side will display the configured geometric dimensions. An explanation of the transport equation used in the two-dimensional scenario will also be provided in the lower right-hand corner.



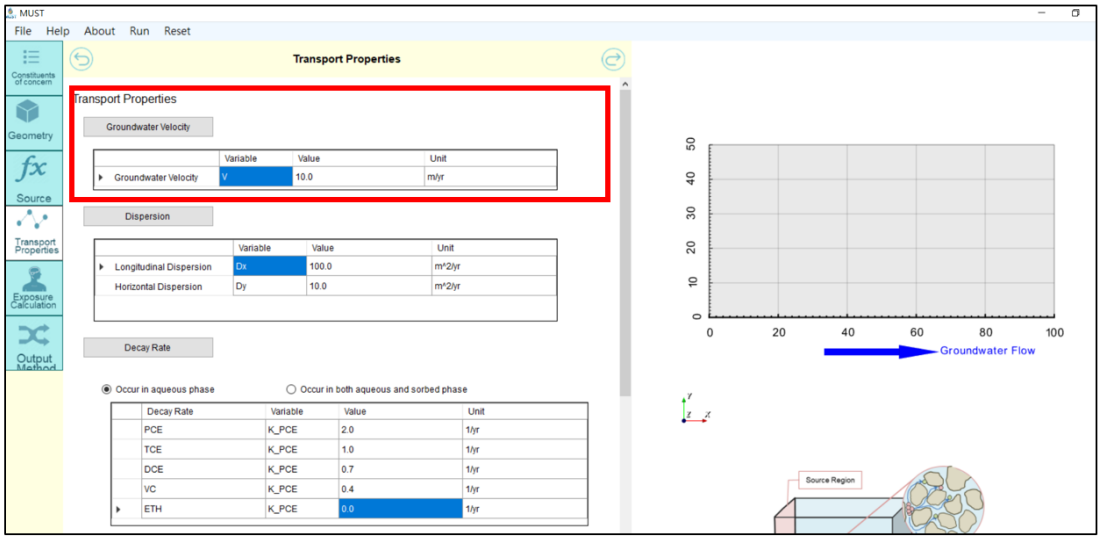
In the third step, you will configure parameters related to contamination sources, including the number of sources, their locations, sizes, and the source concentrations of different contaminants. To begin, you need to determine the number of sources along the inflow boundary (x=0). In this example, we will start with one source. It is necessary to define the position and size of this contaminated source. In this two-dimensional example, the source area can be described by setting two parameters, Y1 and Y2. The interactive screen on the right will provide a visual representation, immediately displaying the dimensions and location of the currently defined source area.



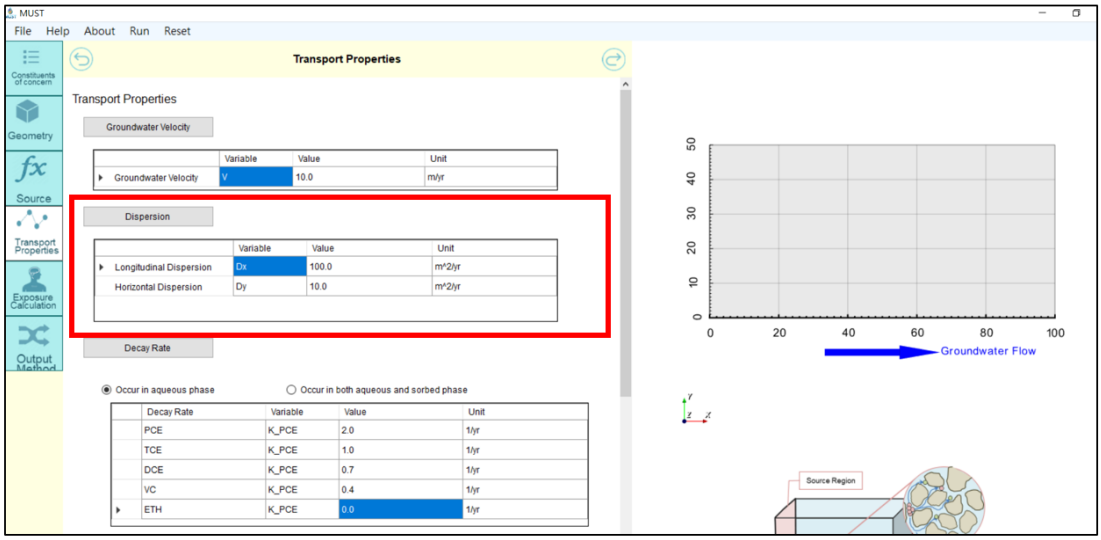
Once the source region is defined, it is necessary to choose the types of inlet boundary conditions and source functions based on changes in the concentration of each contaminant over time. The concentration trend diagram for the selected function, as specified by the user, will be displayed on the right-hand side of the screen. In this particular example, the source continuously releases contaminants at a constant level. The concentration for each contaminant at the source should also be specified.



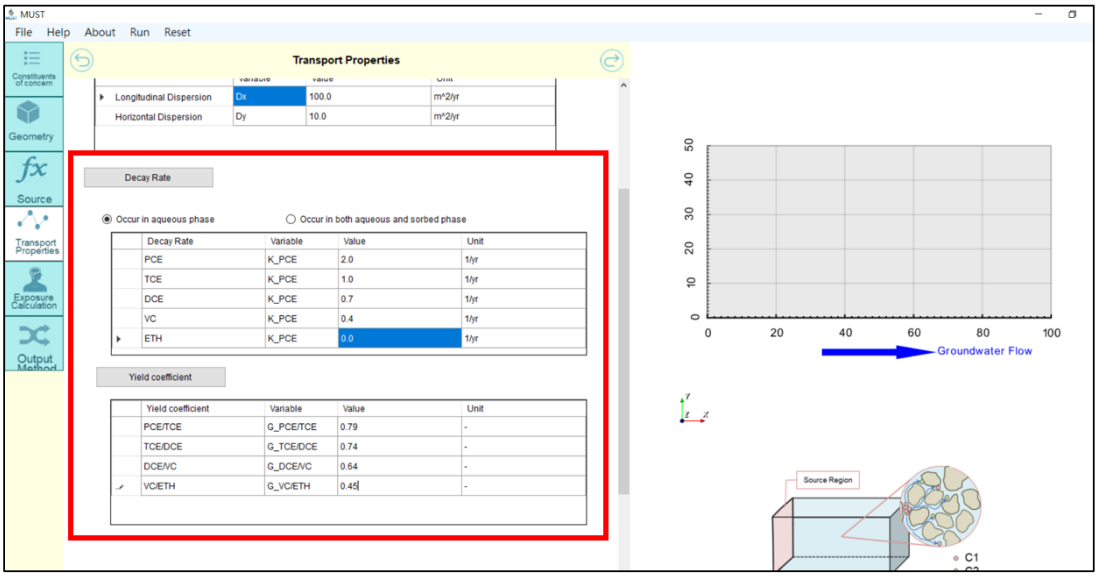
In the fourth step, the transport parameters are input, which include the groundwater flow velocity, dispersion coefficients, and reaction parameters for the contaminants. The groundwater flow velocity represents the average seepage speed within the aquifer. With the input of this parameter, a schematic diagram illustrating the groundwater flow situation will simultaneously appear on the right-hand side of the interface.



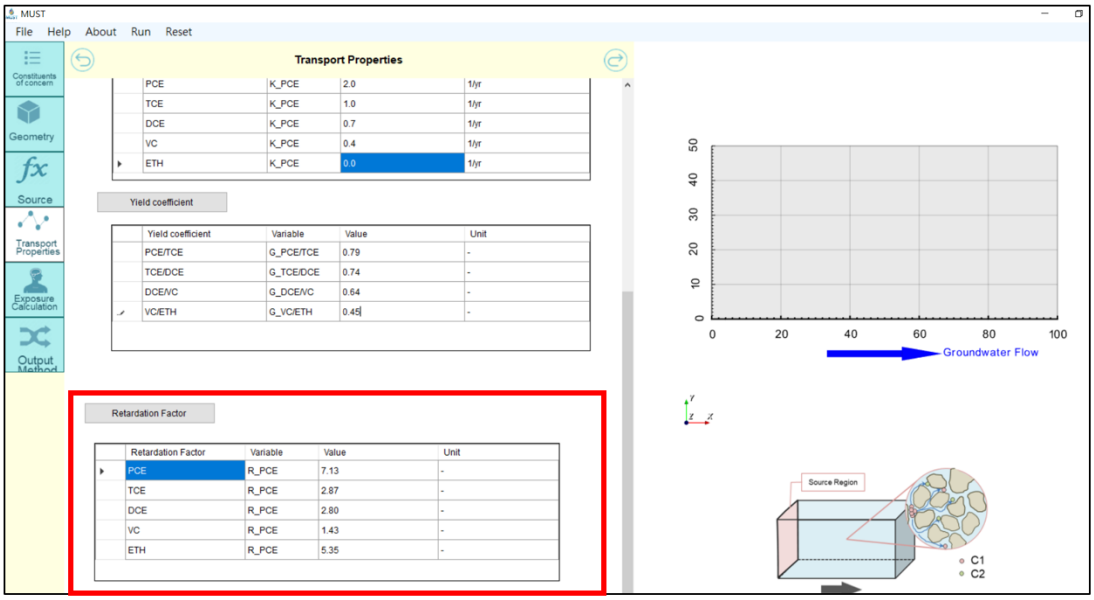
The subsequent step involves inputting the dispersion coefficient. The user needs to provide two dispersion coefficients, one for the longitudinal direction and another for the transverse direction.



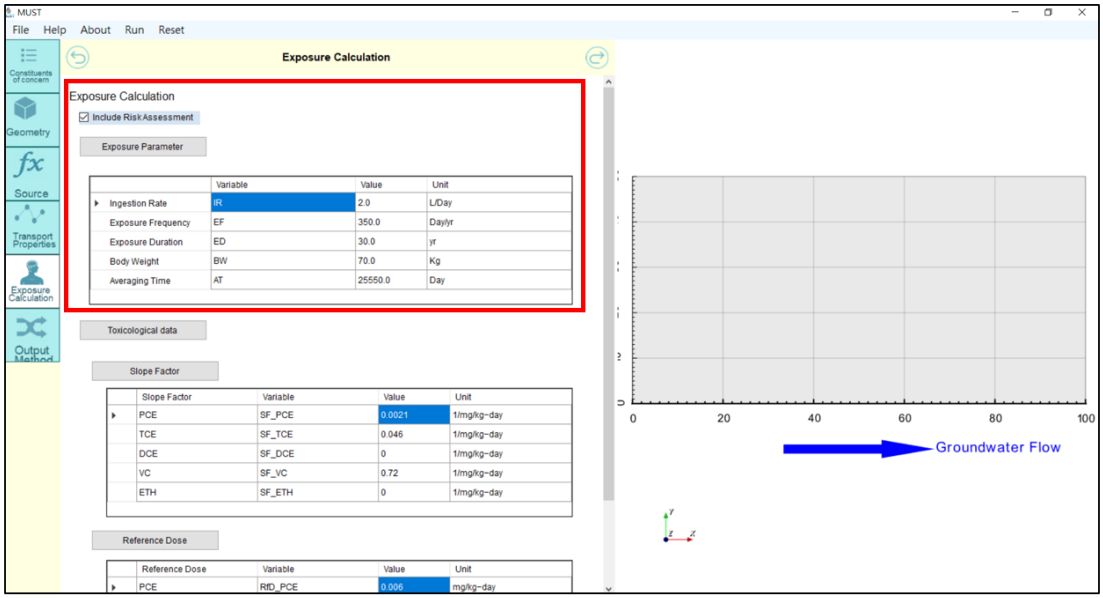
Following that, the degradation or decay rate and yield coefficient need to be addressed.



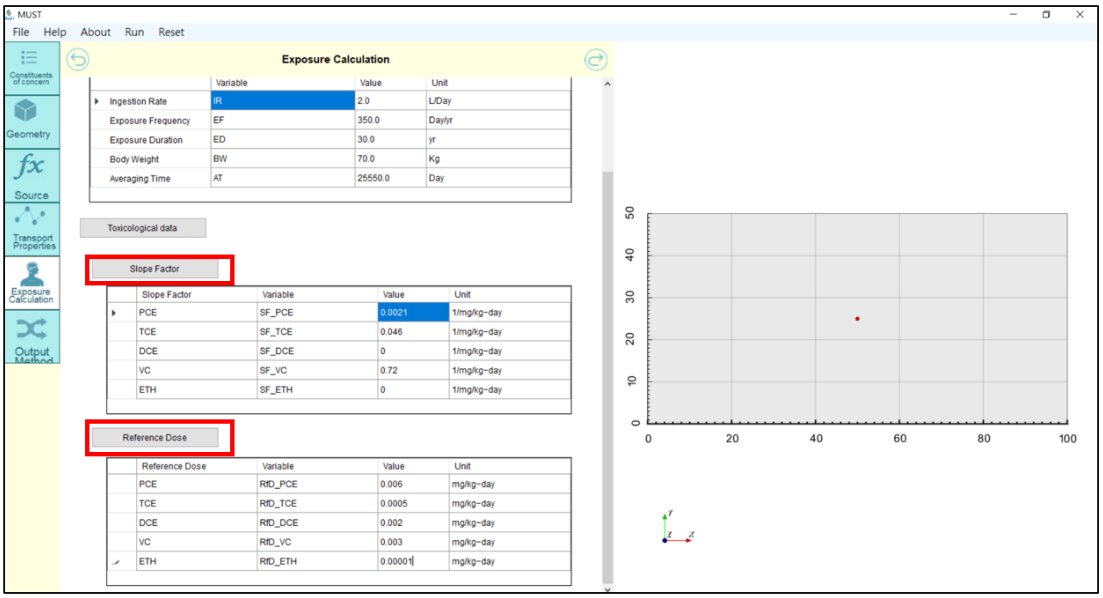
Next, enter the retardation factor, which is associated with the sorption of contaminants. In this software model, it is possible to apply different retardation factors for each contaminant.



In the fifth parameter input step, users have the option to decide whether to integrate the health risk assessment module or not. If the example only requires concentration calculations, this step can be bypassed. If the user desires to calculate the health risks, they should check the “Include Risk Assessment” option. A form for inputting risk-related parameters will be displayed. The upper table in the form presents the exposure parameters related to the human body, such as water intake rate, duration of exposure, body weight, and so on.



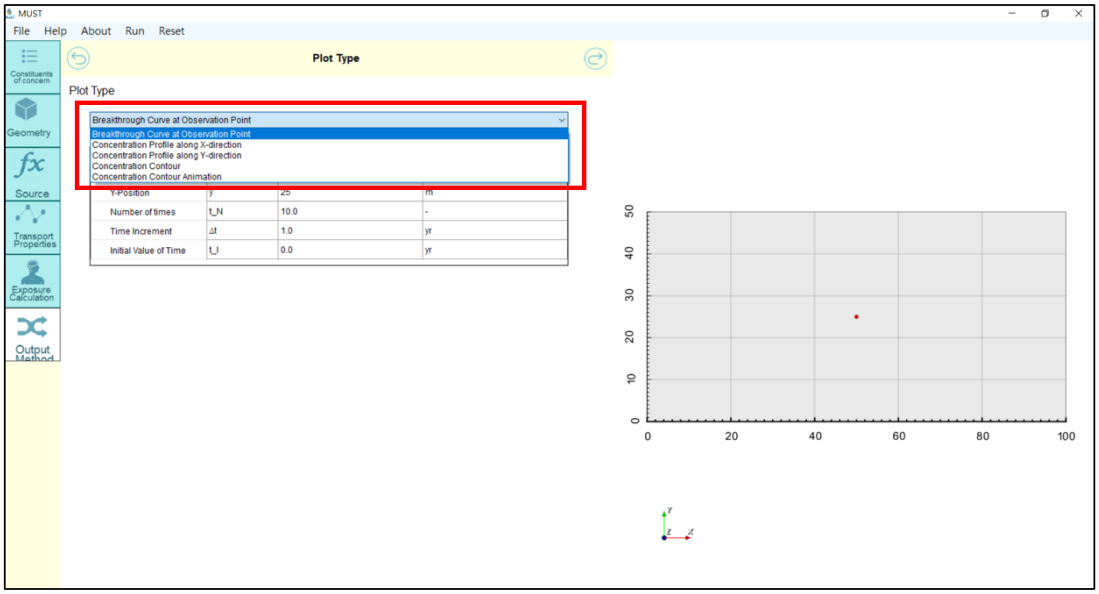
In the subsequent table, input the toxicity parameters for different contaminants. Thees categorized into two groups: the slope factor for carcinogenic risk and the reference dose for non-carcinogenic risk.



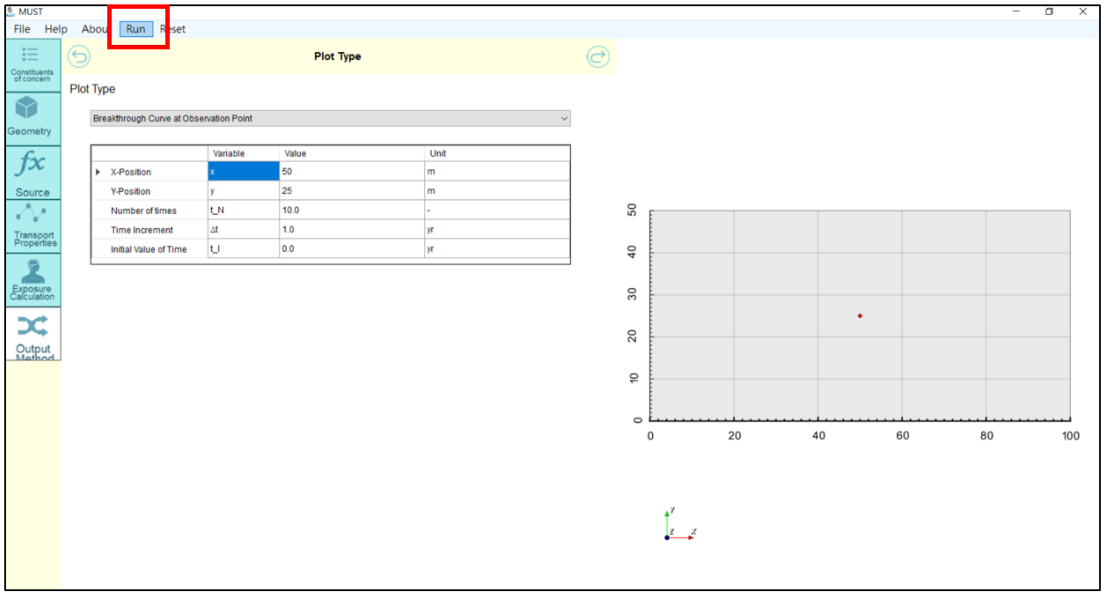
Following the completion of the five parameter input steps, users will proceed to the final step, “Output Method.” In this step, users can select the method of visualizing the output results they wish to present. Detailed options and parameter input for output visualization will be explained in the next chapter.

4. User interface output

The software offers five different methods for visualization of the two-dimensional results. To display changes in concentration at specific observation points over time, the user should choose “Breakthrough Curve at Observation Point” from the dropdown menu under Plot Type. After selecting this option, input the “X-position” and “Y-position” to define the (x, y) coordinate location. A red dot will appear in the interactive area on the right indicating the current simulation calculated position. The “Initial time value” represents the starting time for the simulation calculation, and the total simulation time is determined by the “Number of times” and the “Time increment.”

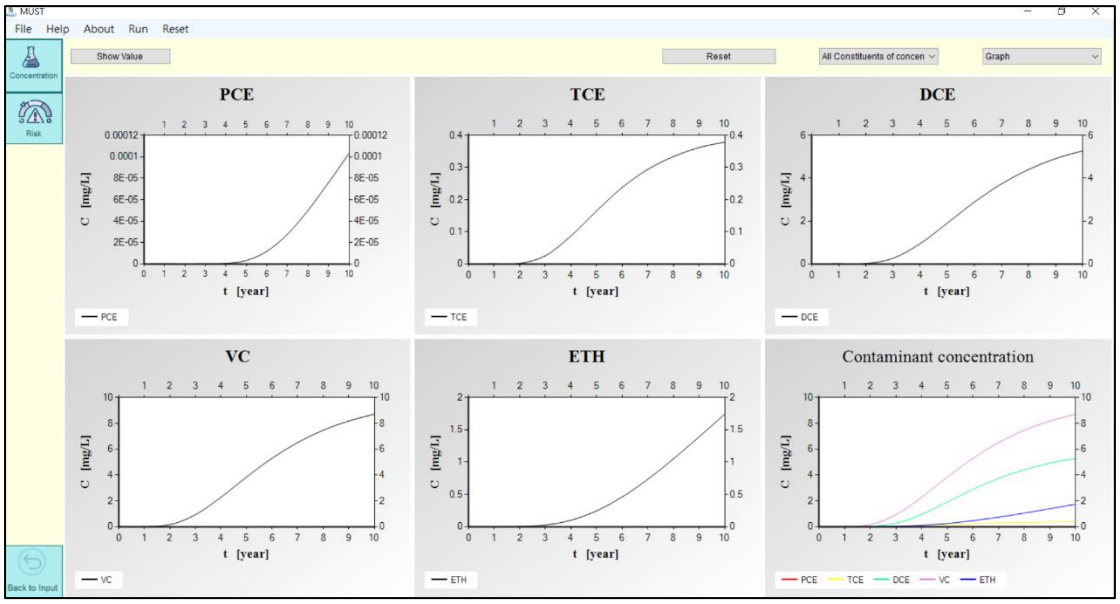


After completing the parameter input related to the desired visualization results, click on the execution button labelled “Run”.

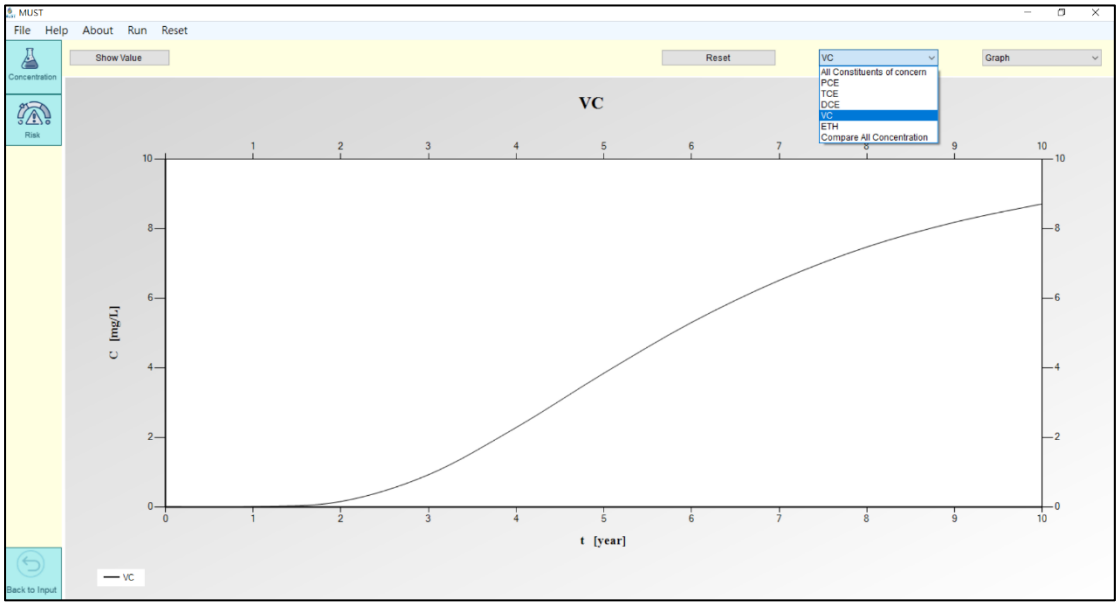


Once the model calculation is finished, a confirmation window will appear. Click

“Confirm” to transition to the subsequent simulation results page. This page will simultaneously display the simulation results for all contaminants specified within this example. In the lower corner, you will find an image that superimposes the simulation results of all contaminants, making it easier to compare their differences.

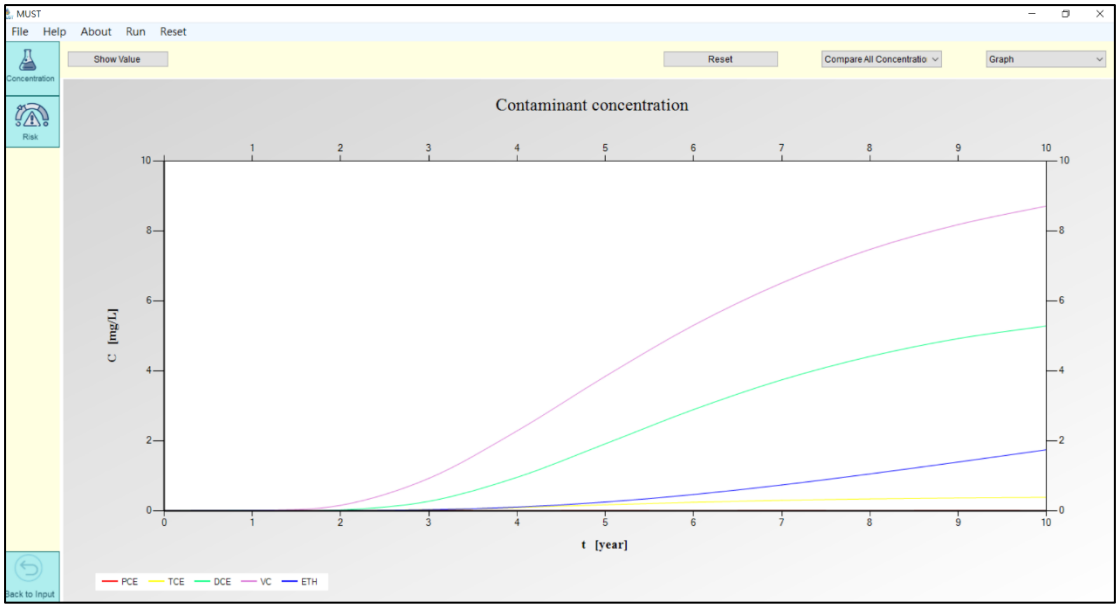


In addition, there is a drop-down menu located in the upper right-hand corner which provides the option for the user to toggle and view the simulation graph for a specific contaminant, for example, to view the simulation results for vinyl chloride exclusively.

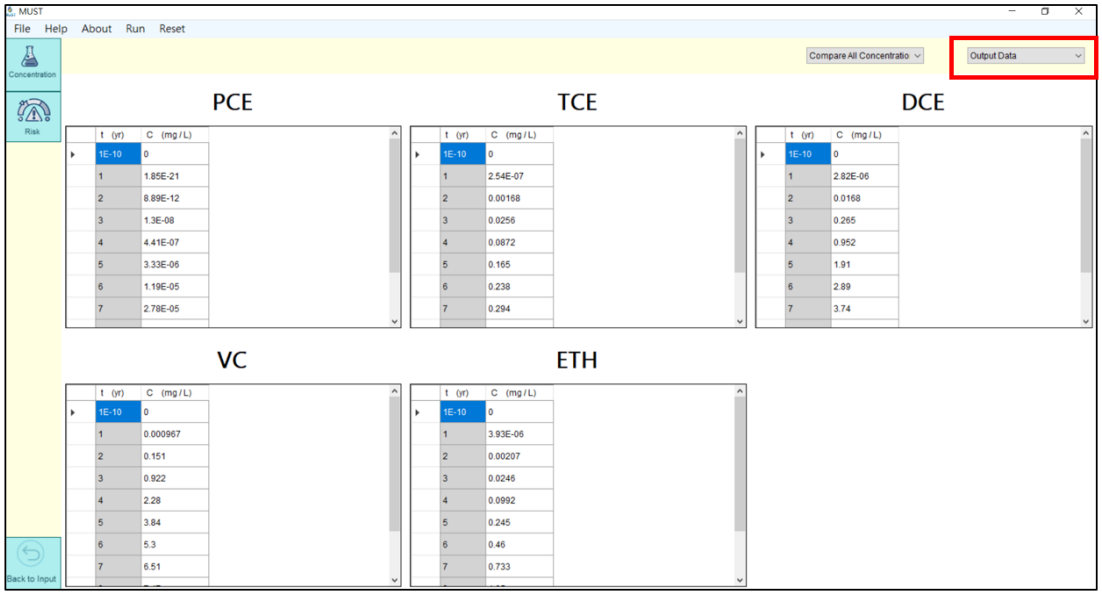


Alternatively, users can choose to display an overlay of the simulated curves for all

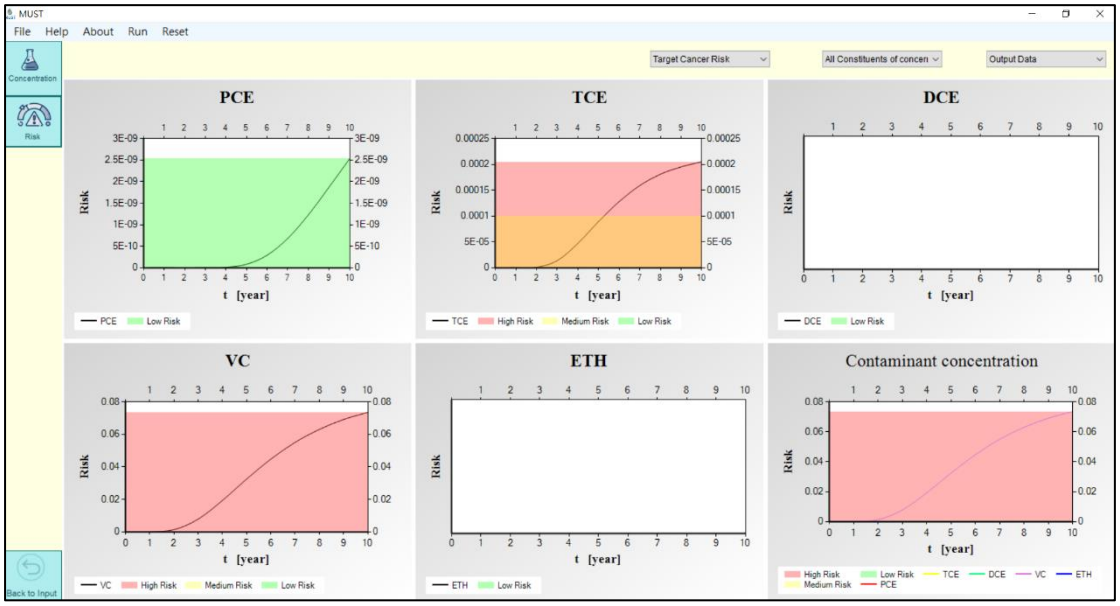
contaminants.



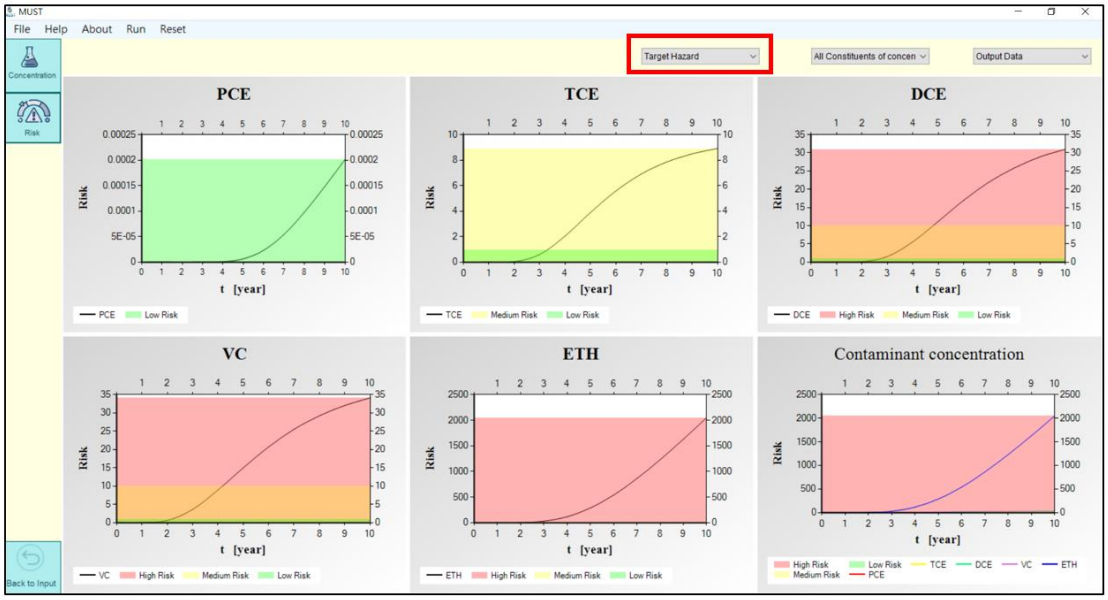
Moreover, the drop-down menu in the upper right-hand corner permits users to switch to a table view, where they can access a detailed list of concentration simulation values at various time points.



If the user wishes to include the health risk module, they should click on the “Risk” button on the left to transition from the concentration calculation results to health risk quantification results. In this view, the results for all contaminants will be displayed simultaneously, color-coded in green, orange, and red signifying low-, medium-, and high-risk levels, respectively.



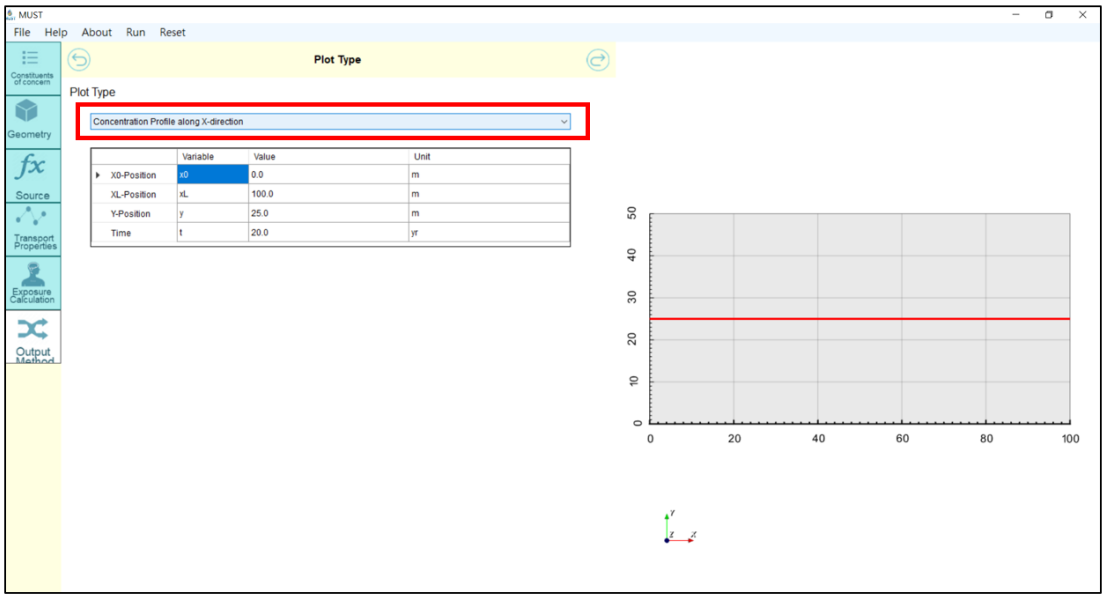
The carcinogenic or non-carcinogenic risk can be toggled by using the drop-down menu located within the red-framed section in the figure.



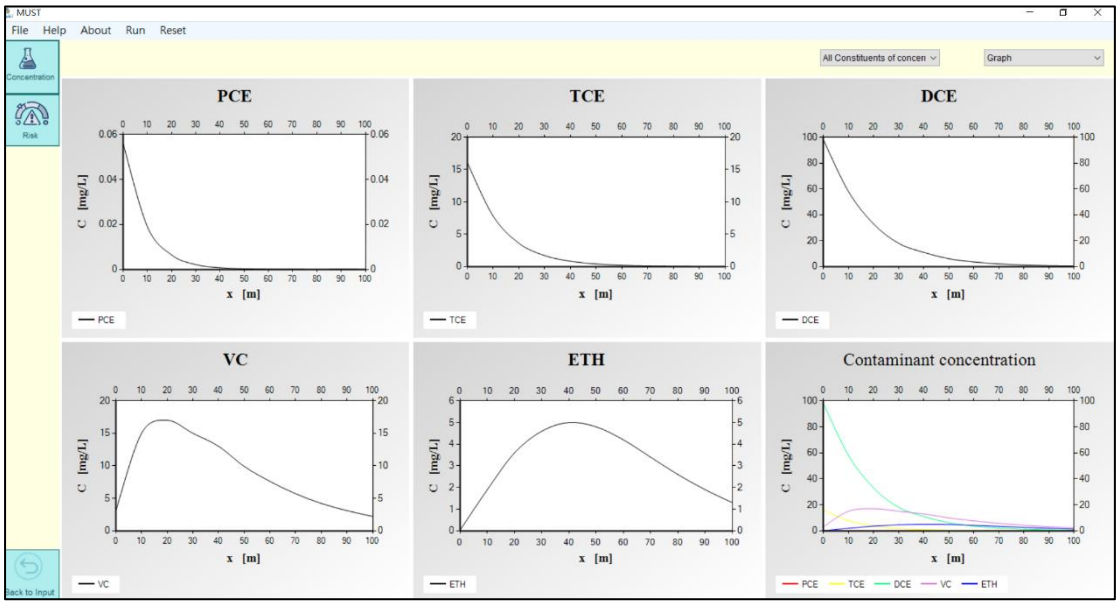
When finished, to return to the input parameter page, simply click the “Back to input” button located in the lower-left corner.

To simulate the concentration profile at various distances in the direction of the flow,

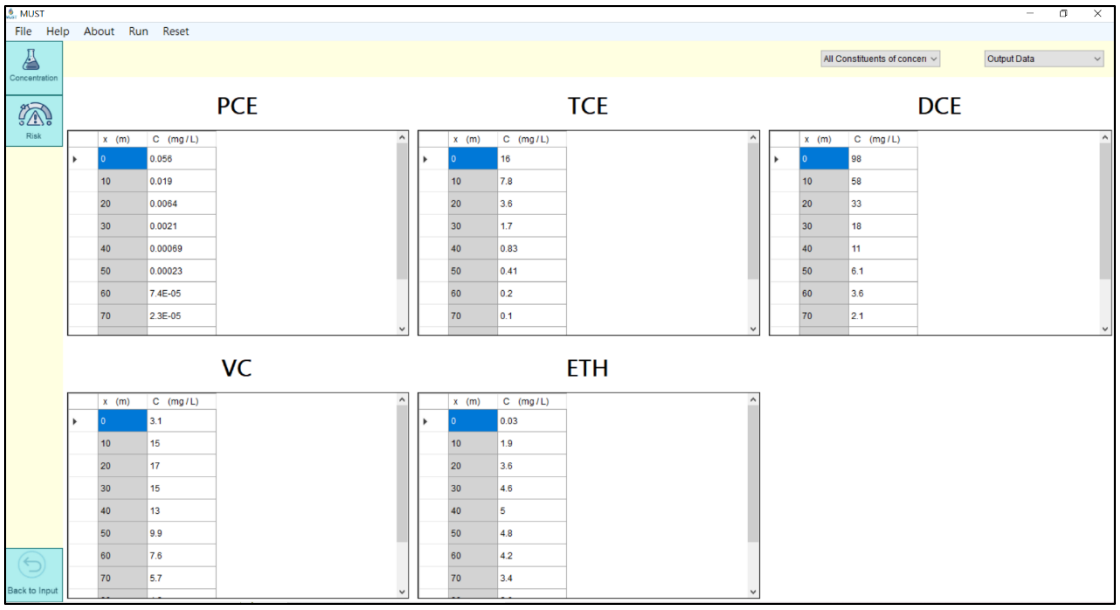
select the option “Concentration profile along the x-direction” on the output option page, then to input the “X0-Position,” “XL-Position,” and “Y Position” to define the position and distance along a straight line. The schematic diagram on the right will display the simulation results for the currently defined section as a red line. For instance, this line might represent the position at the center of the source zone (y=25 m) along the x-direction from 0 to 100 m. Lastly, enter the simulation time in the field labeled “Time.”



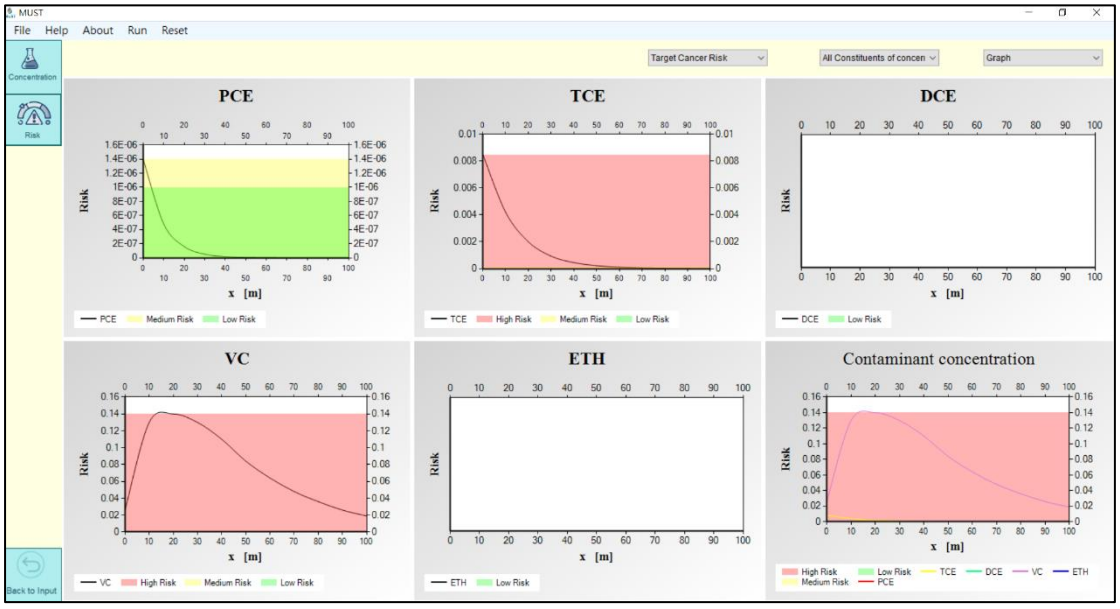
Similarly, after clicking the execute button, the results showing the concentrations profiles of the different contaminants along with the distance will be displayed.



The concentration values at various observation points can also be displayed in a table.

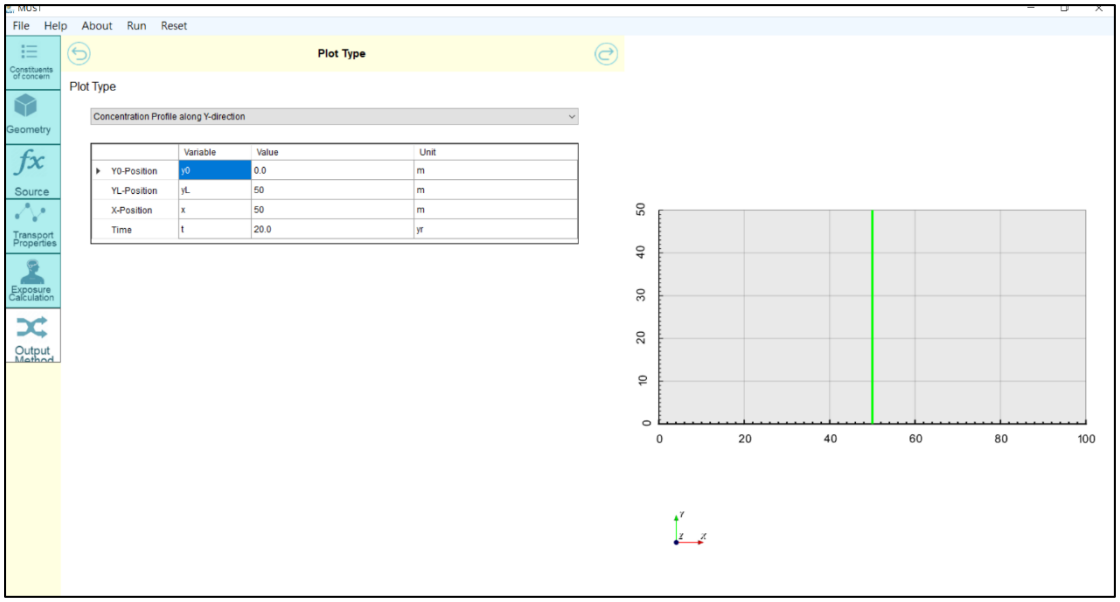


To display the risk profile for these simulation results, click the “Risk” button on the left.

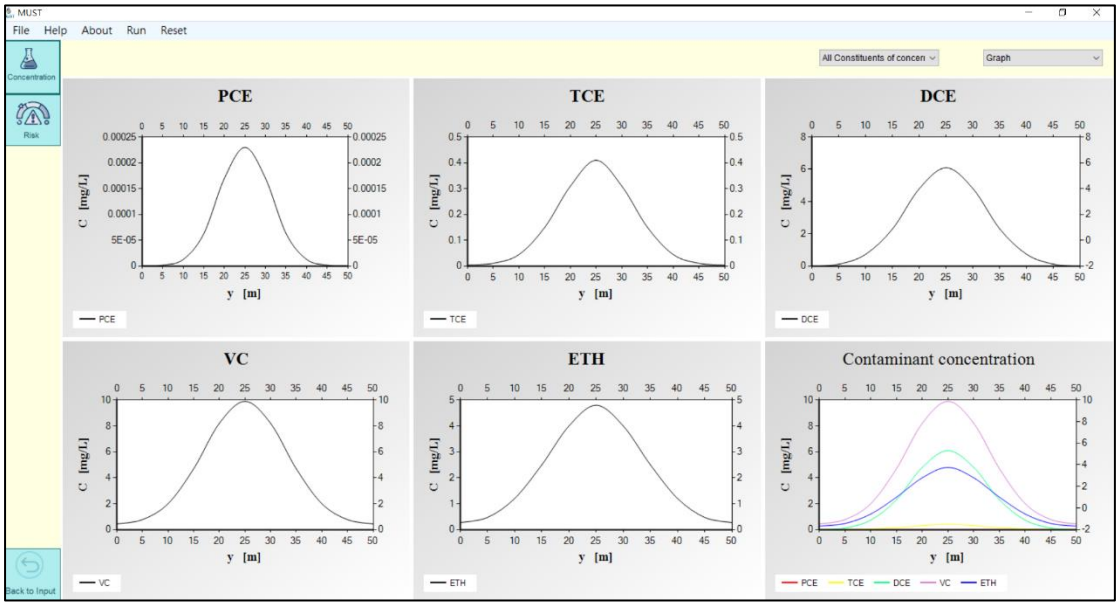


With the visualization of the two-dimensional transport output results, there is an additional option called “Concentration profile along the y-direction,” for displaying the concentration profiles along the y direction. The input parameters are analogous to

those described previously. For example, x=50 m represents the distance from 0 to 50 meters in the y direction. The simulated distance is depicted in the schematic diagram on the right by a green line.

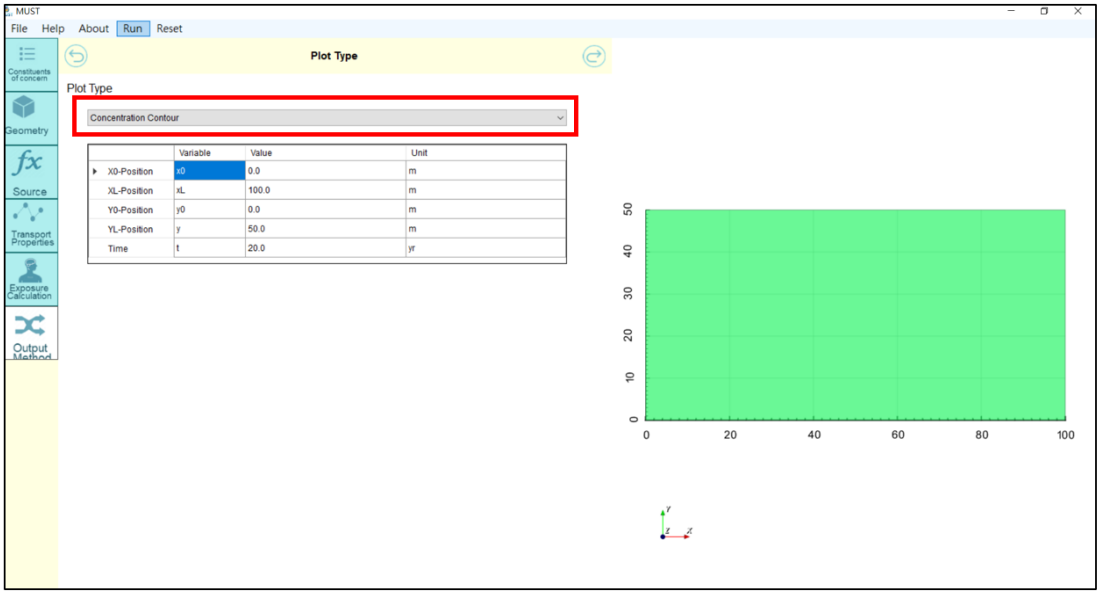


These simulation results provide an understanding of the concentration profiles perpendicular to the groundwater flow direction.

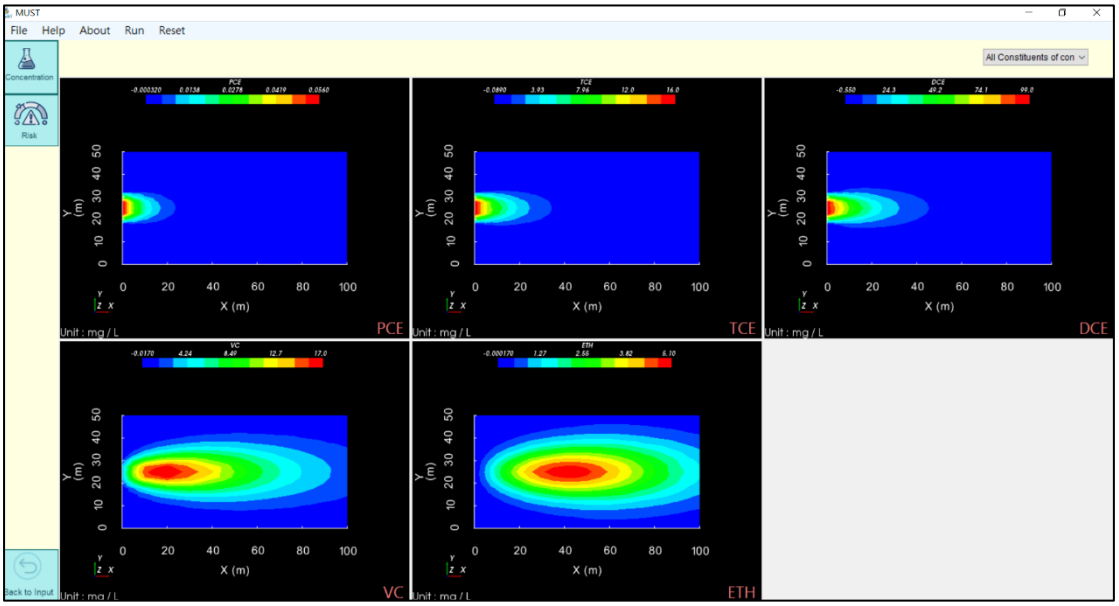


To simulate the distribution of the plume on a plane, the user can select “Concentration contour” in the output options. For this two-dimensional scenario, the user only needs to input four parameters: “X0-Position,” “XL-Position,” “Y0-Position,” and “YL-

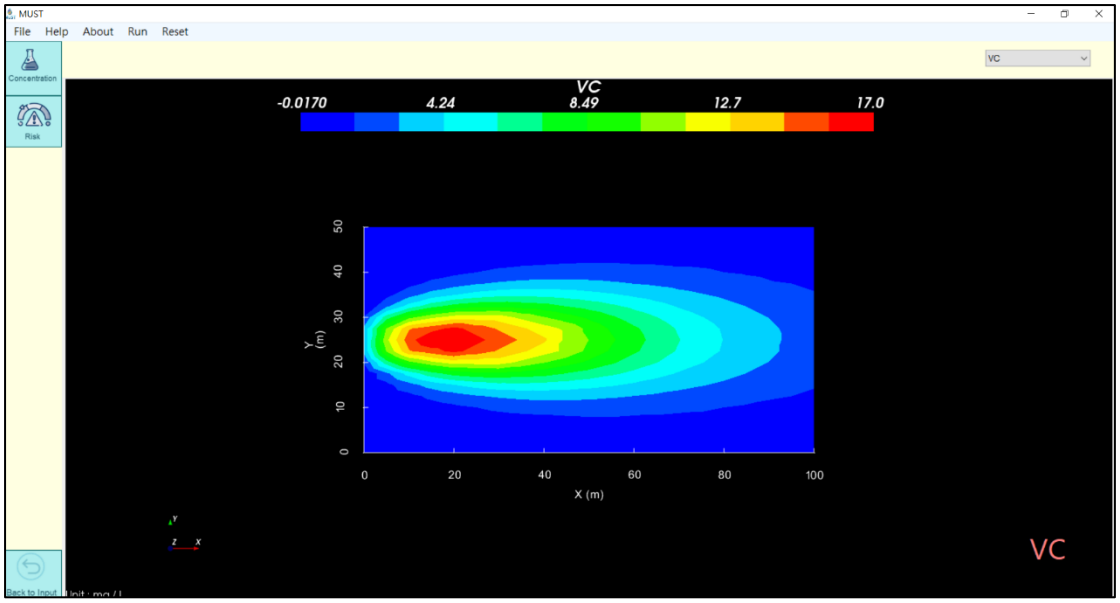
Position.” These parameters define the area to be simulated. The three-dimensional scenario requires an additional parameter, the depth in the vertical direction as input. The green area shown in the diagram on the right represents the range defined by the user. Finally, the user enters the time the want to simulate.



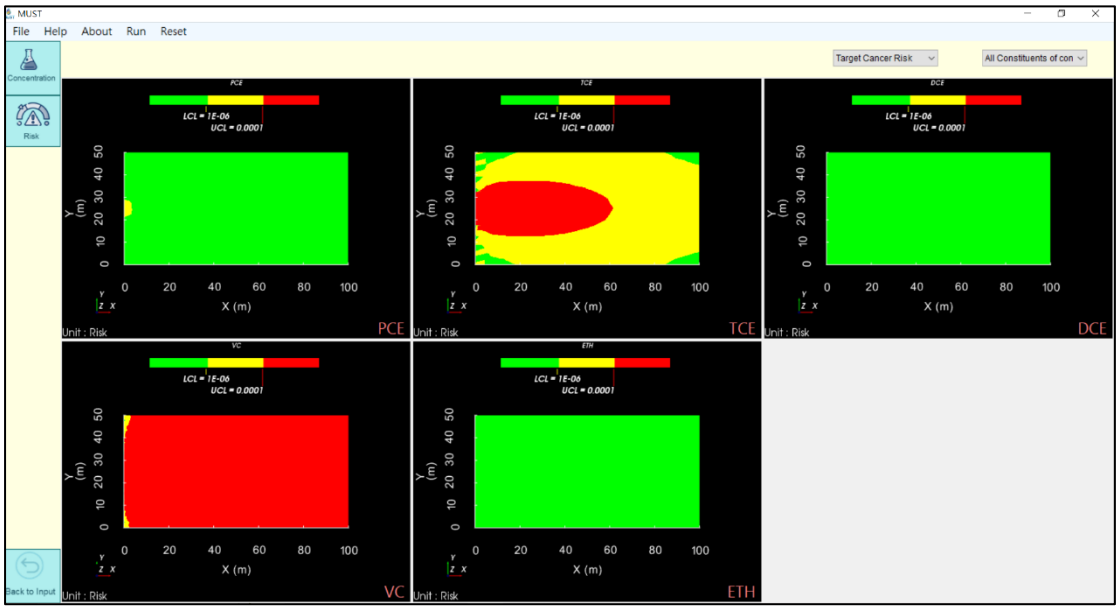
After clicking “Execute,” the software will display the migration and distribution of the different contaminants, distinguishing concentration levels through color-coding.



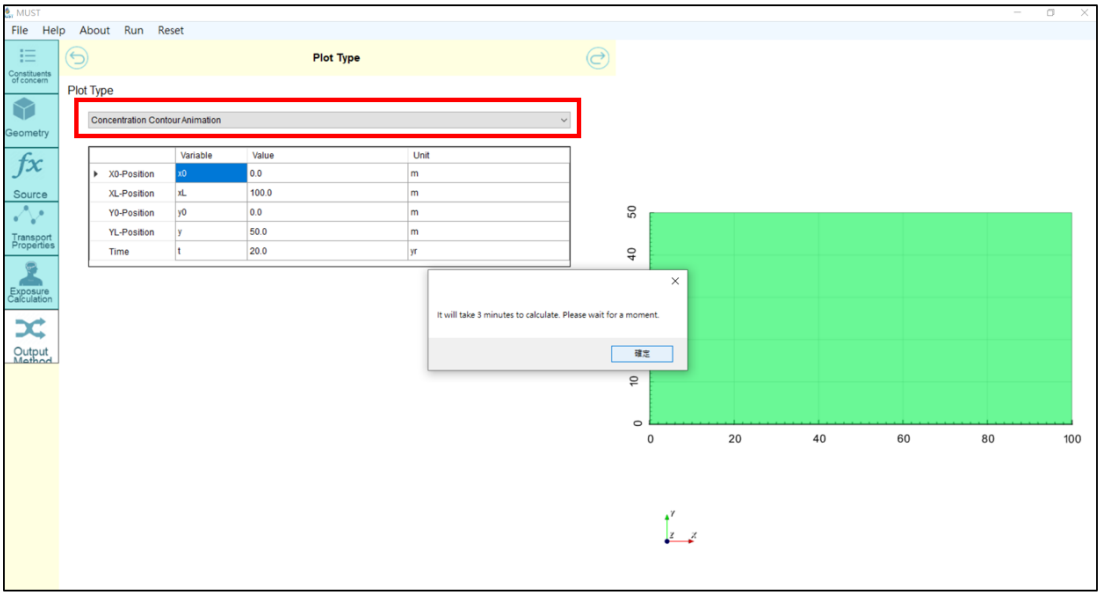
The user can also switch to focus solely on the representation of a specific contaminant.



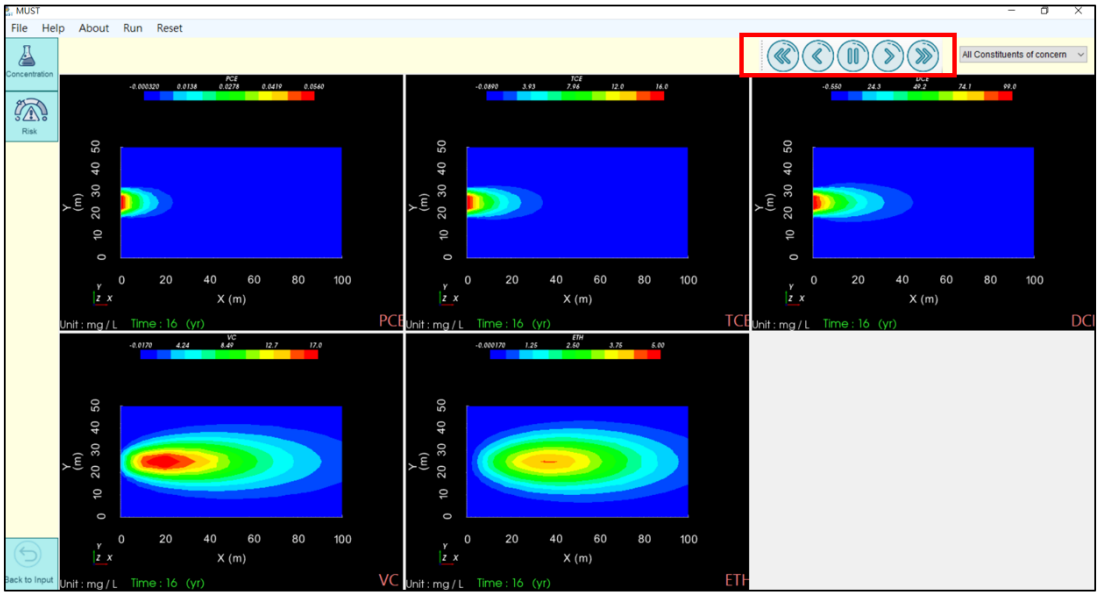
The “Risk” button to the left can be used to display the spatial distribution results for health risk quantification, which makes it easier to identify high-risk areas.



Additionally, the software will display an animation of the plume migration over time, by clicking “Concentration Contour Animation” on the output options page. The results will automatically transition through five time periods, according to the input simulation times.



After executing and completing the calculation, the migration animation will be automatically played. The buttons above can be used to pause, fast forward, or rewind to display as desired.



References

Aziz, C.E., Newell, C.J., Gonzales, J.R., Hass P., Clement, T.P., Sun, Y., 2000. BIOCHLOR-Natural attenuation decision support system v1.0, User's Manual. US EPA Report, EPA 600/R-00/008.

Falta, R.W., Stacy, M.B., Ahsanuzzaman, W.M., Earle, R.C., 2007. Remediation Evaluation Model for Chlorinated Solvents (REMChlor), EPA/600/C-08/001.

Funk, S.P., Hnatyshin, D., Alessi, D.S., 2017. HYDROSCAPE: A new versatile software program for evaluating contaminant transport in groundwater. SoftwareX 6:261–266. https://doi.org/10.1016/j.softx.2017.10.001.

Liao, Z.Y., Suk, H., Liu, C.W., Liang, C.P., Chen, J.S., 2021. Exact analytical solutions with great computational efficiency to three-dimensional multispecies advection-dispersion equations coupled with a sequential first-order degradation reaction network. Adv. Water Resour. 155, 104018. https://doi.org/10.1016/j.advwatres.2021.104018.

Moranda, A., Cianci, R., Paladino, O., 2018. Analytical Solutions of One-Dimensional Contaminant Transport in Soils with Source Production-Decay. Soil Syst. 2018, 2(40) https://doi.org/10.3390/soilsystems2030040.

Newell, C.J., R.K. McLeod, and J.R. Gonzales. 1996. BIOSCREEN Natural Attenuation Decision Support System User's Manual Version 1.3, USEPA/600/R-96/087. Washington, DC: U.S. USEPA National Risk Management Research Laboratory.

Wang, X., Neville, C.J., 2019. A semi-analytical solution for the transport of solutes with complex sequences of first-order reactions. Comput. Geosci. 123, 121–136. https://doi.org/10.1016/j.cageo.2018.11.013.

Yeh, G.T. 1981. AT123D: Analytical Transient One-, Two-, and Three-Dimensional Simulation of Waste Transport in the Aquifer System. ORNL-5602, Oak Ridge National Laboratory.

Appendix A. Introduction to the analytical model

The model used in MUSt software is based on the three-dimensional multispecies transport analytical model developed by Liao et al. (2021). The geometry of the groundwater system considered in the model is shown in Figure A1, where L is the length in the x direction; W is the width in the y direction; H is the height in the z direction. The red area is the location of the source. The groundwater flow is assumed to be steady and uniform along the x direction.

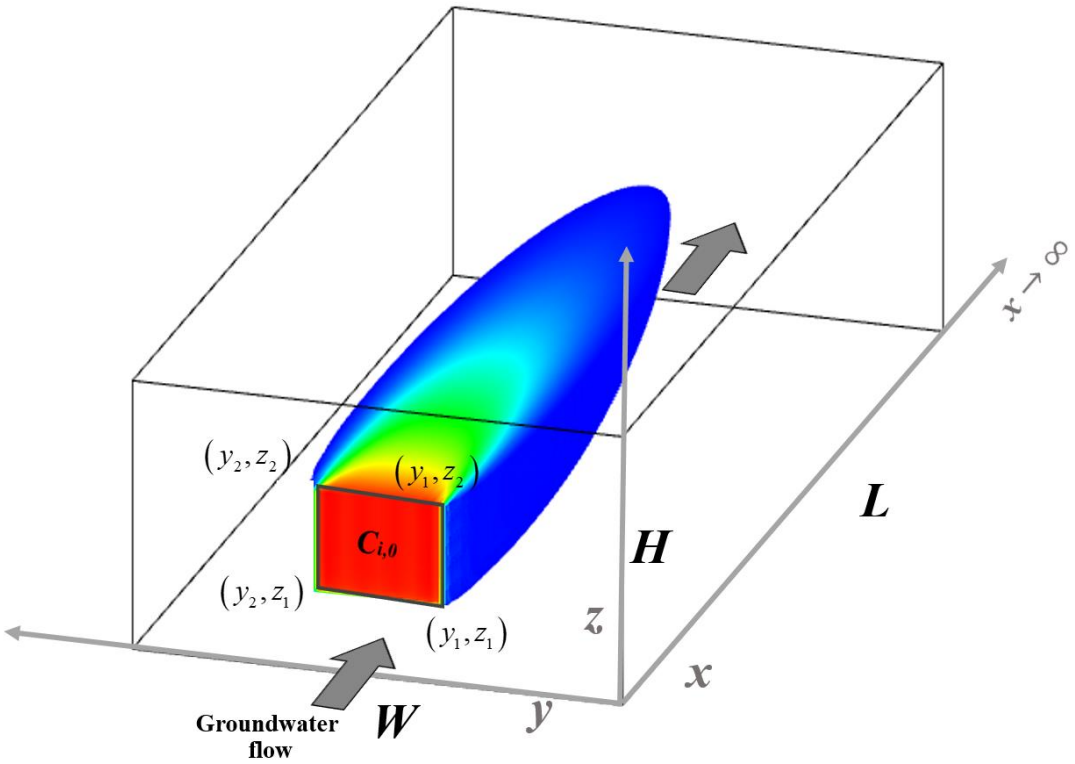


Figure A1. Schematic diagram of multispecies transport in the simulated system.

A.1 Governing equations for solute transport

This model mainly solves a set of partial differential equations describing the transport of five contaminants with a sequential degradation in the groundwater system. The main transport reaction mechanisms considered include one-dimensional advection

and three-dimensional dispersion, as well as linear isothermal equilibrium sorption and sequential first-order degradation. The relevant equations for describing different contaminants are as follows:

$$D_x \frac{\partial^2 C_1(x,y,z,t)}{\partial x^2} + D_y \frac{\partial^2 C_1(x,y,z,t)}{\partial y^2} + D_z \frac{\partial^2 C_1(x,y,z,t)}{\partial z^2} - v\frac{\partial C_1(x,y,z,t)}{\partial x} - \mu_1 C_1(x,y,z,t) = R_1 \frac{\partial C_1(x,y,z,t)}{\partial t} \quad ; \quad (1)$$

$$D_x \frac{\partial^2 C_2(x,y,z,t)}{\partial x^2} + D_y \frac{\partial^2 C_2(x,y,z,t)}{\partial y^2} + D_z \frac{\partial^2 C_2(x,y,z,t)}{\partial z^2} - v\frac{\partial C_2(x,y,z,t)}{\partial x} + g_{1\to 2}\mu_1 C_1(x,y,z,t) - \mu_2 C_2(x,y,z,t) = R_2 \frac{\partial C_2(x,y,z,t)}{\partial t} \quad ; \quad (2)$$

$$D_x \frac{\partial^2 C_3(x,y,z,t)}{\partial x^2} + D_y \frac{\partial^2 C_3(x,y,z,t)}{\partial y^2} + D_z \frac{\partial^2 C_3(x,y,z,t)}{\partial z^2} - v\frac{\partial C_3(x,y,z,t)}{\partial x} + g_{2\to 3}\mu_2 C_2(x,y,z,t) - \mu_3 C_3(x,y,z,t) = R_3 \frac{\partial C_3(x,y,z,t)}{\partial t} \quad ; \quad (3)$$

$$D_x \frac{\partial^2 C_4(x,y,z,t)}{\partial x^2} + D_y \frac{\partial^2 C_4(x,y,z,t)}{\partial y^2} + D_z \frac{\partial^2 C_4(x,y,z,t)}{\partial z^2} - v\frac{\partial C_4(x,y,z,t)}{\partial x} + g_{3\to 4}\mu_3 C_3(x,y,z,t) - \mu_4 C_4(x,y,z,t) = R_4 \frac{\partial C_4(x,y,z,t)}{\partial t} \quad ; \quad (4)$$

$$D_x \frac{\partial^2 C_5(x,y,z,t)}{\partial x^2} + D_y \frac{\partial^2 C_5(x,y,z,t)}{\partial y^2} + D_z \frac{\partial^2 C_5(x,y,z,t)}{\partial z^2} - v\frac{\partial C_5(x,y,z,t)}{\partial x} + g_{4\to 5}\mu_4 C_4(x,y,z,t) - \mu_5 C_5(x,y,z,t) = R_5 \frac{\partial C_5(x,y,z,t)}{\partial t} \quad . \quad (5)$$

Here, $C_i(x,y,z,t)$ is the solute concentration in the dissolved phase [M/ L^3]; x, y, z are spatial coordinates [L]; t is the time [T]; D_x is the longitudinal dispersion coefficient in the x direction [L^2/T]; D_y and D_z are the horizontal and vertical transverse dispersion coefficients [L^2/T]; v is the average groundwater seepage velocity [L/T]; μ_i is the first-order decay reaction rate of the ith species [1/T]; $g_{i-1\to i}$ is the yield coefficient [-]; R_i is the retardation factor of the ith species [-].

A.2 Initial and boundary conditions

The groundwater system is assumed to be free of any contaminant mass, for each contaminant, at the initial time:

$$C_i(x,y,z,t=0)=0 \qquad i=1,....,5\,. \tag{A1}$$

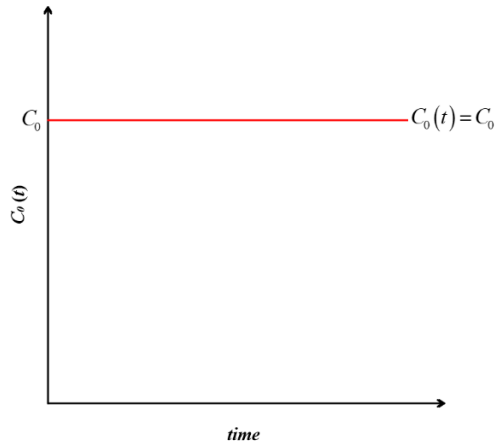
At the source, two types of inlet boundary conditions (BCs) are considered, the first-type and the third-type, as shown in Eq. (A2). T source concentration may change with time, requiring further derivation of new solutions with different source functions. This model, which is based on the three-dimensional model developed by Liao et al. (2021), can not only consider a constant source concentration, but it can also consider the time-varying functions of exponential decay and the arbitrary concentration at different times, according to the field data. Thus, there are three different source functions and the change in the concentration at the source with time, as shown in Figure A2.

$$C_i(x=0,y,z,t)=f_{i,0}(t) \qquad y_1 \le y \le y_2 \quad z_1 \le z \le z_2$$

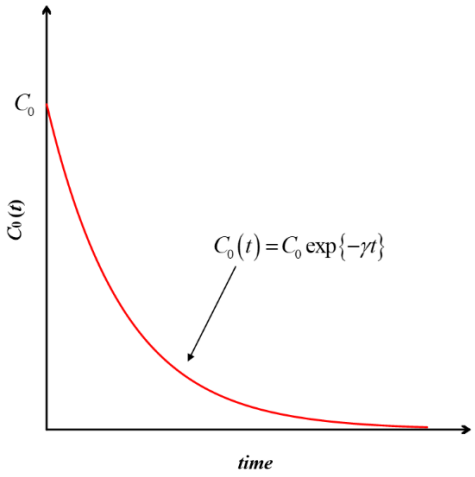
$$i=1,....,5 \qquad \text{for the first-type BC} \tag{A2a}$$

$$-D_x\frac{\partial C_i(x=0,y,z,t)}{\partial x}+vC_i(x=0,y,z,t)=vf_{i,0}(t) \qquad y_1 \le y \le y_2 \quad z_1 \le z \le z_2$$

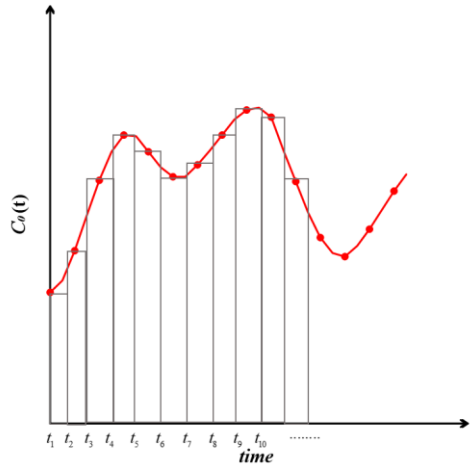
$$i=1,....,5 \qquad \text{for the third-type BC} \tag{A2b}$$



(a) Constant concentration



(b) Exponential decay



(c) Piecewise constant concentration

Figure A2. Schematic diagram showing the three source functions. The vertical axis gives the concentration value and the horizontal axis indicates the time.

The three source functions considered $f_{i,0}(t)$ can be specified as follows:

Constant concentration

$$f_{i,0}(t) = C_{i,0} \qquad i = 1,....,5\,; \tag{A3}$$

Exponential decay

$$f_{i,0}(t) = C_{i,0}\, exp(-\gamma_i t) \qquad i = 1,....,5\,, \tag{A4}$$

where γ is the source decay rate constant [1/T].

Piecewise constant concentration

$$f_{i,0}(t) = \sum_{j=1}^{j=N} \mathrm{C}_{i,j}\left[H(t - t_{j-1}) - H(t - t_j)\right] \qquad i = 1,....,5 \tag{A5}$$

which can approximately represent the observed concentration $C_{i,j} = C_{i,1}, C_{i,2}, C_{i,3}, ..., C_{i,N}$ at different times $t = 0, t_1, t_2, t_3, ..., t_N$ using multiple unit step functions.

The exit boundary conditions are specified at infinity as follows:

$$C_i(x \to \infty, y, z, t) = 0 \quad i = 1,....,5\,. \tag{A6}$$

It is assumed there will be no contaminant flux moving across the right and left boundary plane in the *y* direction or on the upper and lower boundary plane in the *z* direction. The other four boundary conditions are denoted as follows:

$$\frac{\partial C_i(x, y = 0, z, t)}{\partial y} = 0 \qquad i = 1,....,5 \tag{A7}$$

$$\frac{\partial C_i(x, y = W, z, t)}{\partial y} = 0 \qquad i = 1,....,5 \tag{A8}$$

$$\frac{\partial C_i(x, y, z = 0, t)}{\partial z} = 0 \qquad i = 1,....,5 \tag{A9}$$

$$\frac{\partial C_i(x, y, z = H, t)}{\partial z} = 0 \qquad i = 1,....,5 \tag{A10}$$

The final analytical solutions for each species can be obtained as in Liao et al. (2021).

A.3 Health risk assessment module

This software integrates a health risk assessment module, mainly used to quantify the risks caused by long-term exposure to groundwater contaminants in the aquifer. The health risk quantification method divides the risk into two types: Carcinogenic risk and Non-carcinogenic risk.

(a) Carcinogenic risk estimation

Certain contaminants are considered to be carcinogenic or possibly carcinogenic, and are generally represented by the probability of the target risk (TR), with 10^{-6} as the standard. The calculation formula is as follows:

$$TR = \frac{C \times IR \times EF \times ED}{BW \times AT} \times CSF \ , \tag{A11}$$

where C is the dissolved concentration in the groundwater [mg/L]; IR is intake rate [L/day]; EF is the exposure frequency [day/year]; ED is the exposure duration [year]; BW is the body weight [kg]; AT is the average time [day]; CSF is the slope factor [1/(mg/kg-day)].

(b) Non-Carcinogenic risk estimation

The non-carcinogenic risk is calculated to find the Hazard quotient (HQ) which is used to indicate the risk to the health of the human body caused by harmful non-carcinogenic substances in the environment. The HQ represents the possibility of the occurrence of diseases other than cancer, usually with 1 as the reference standard. The formulation is as follows:

$$HQ = \frac{C \times IR}{BW \times RfD} \ , \tag{A12}$$

where C is dissolved concentration in the groundwater [mg/L]; IR is the intake rate

[L/day]; BW is the body weight [kg]; RfD is the reference dose [mg/kg-day].

The contaminant concentrations are calculated with the software's analytical solute transport model, and the relevant exposure and toxicity parameters input, which are combined in the risk calculation module to quantify the risk.

Appendix B. Simulation Cases

The nine reference cases simulated using the MUSt software are discussed below. In the first step, as shown in Figure B1, a drop-down menu appears where the "Constituents of Concern" are divided into different types of contaminants. Once selected, the interface will automatically import the parameters for each specific case. The user then clicks "Run" to initiate simulation operations to quickly obtain the results. If the input parameters need to be modified, corrections can be made directly for each step. Modifying the parameters will not change the parameters of the original default case. The user needs to save the file of this simulation.

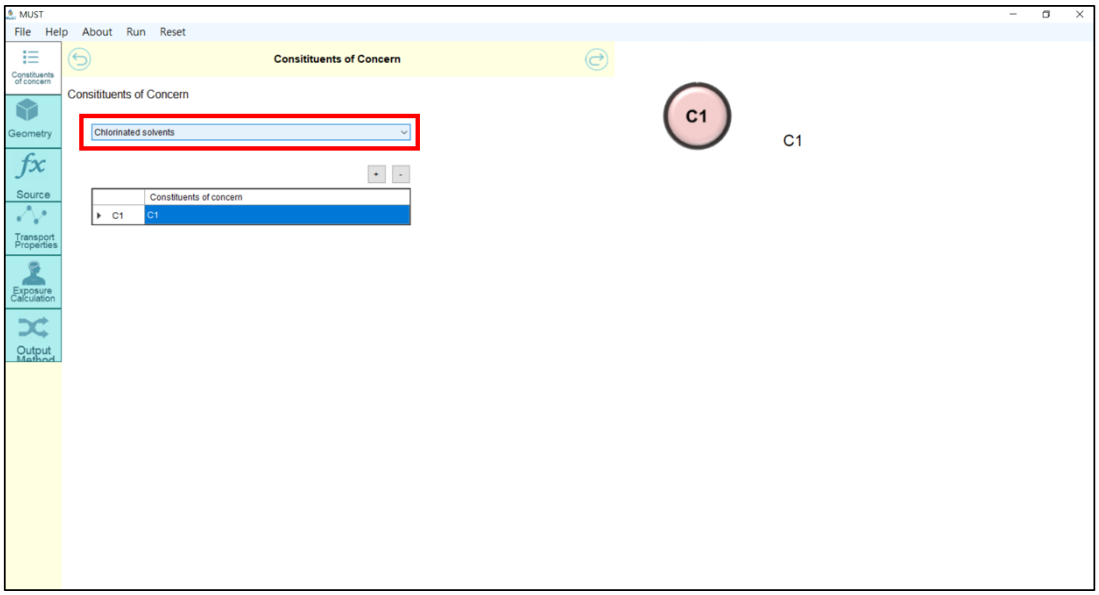


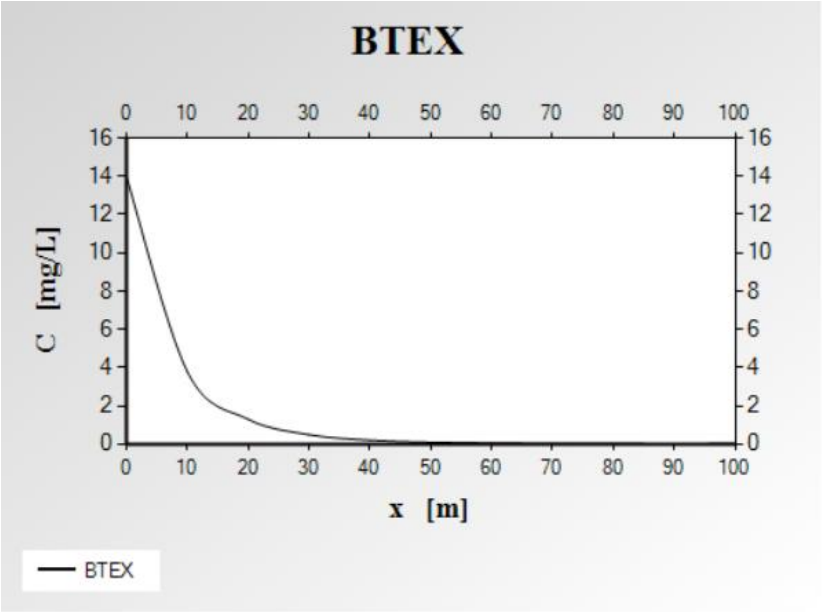
Figure B1. Case selection on the software interface.

Case 1. Single species transport: Benzene, Toluene, Ethylbenzene, Xylene

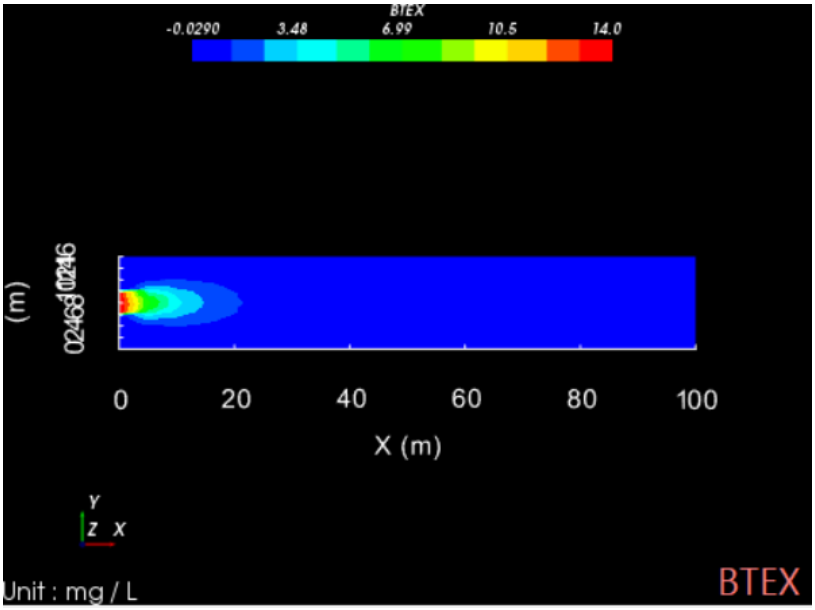
This scenario is from the BIOSCREEN user manual. This case simulates the contamination of a site by the non-aqueous liquid BTEX compounds (Benzene, Toluene, Ethylbenzene, Xylene) that occurred from 1989 to 1995 in the United States. Considering the release of a single species, the relevant geometry of the aquifer, and the reaction and transport parameters are listed in Table B1; the concentration of the source is 13.68 mg/L. The main assumption for this case is that the source is about 4 meters wide and located between $6\ \mathrm{m} \le y \le 10\mathrm{m}$ at x=0. After inputting the relevant parameters, the spatial distribution of the plume can be simulated. Figure B2 shows the concentration distribution along the axis (y=8 m) and the plume migration at 6 years.

Table B1. Parameters for Case 1 (BIOSCREEN user's manual)

Parameters	Value
Length, L [m]	100
Width, W [m]	16
Seepage velocity, v [m year^{-1}]	34.68
Longitudinal dispersion coefficient, D_x [m^2 year^{-1}]	343
Transverse dispersion coefficient, D_y [m^2 year^{-1}]	34.7
Retardation factor, R_1 [-]	1
Decay rate constant, μ_1 [year^{-1}]	4.6
Source concentration, $C_{1,0}$ [mg L^{-1}]	13.68



(a)



(b)

Figure B2. (a) Concentration profile along the axis (y=8 m) at 6 years; (b) plume migration at 6 years.

Case 2. Single species transport: Radionuclide U-234

This case is based on a previous study by Moranda et al. (2018), used to simulate the transport of radionuclides released from a disposal site in the subsurface environment. The contaminant of concern is a single nuclide of Uranium-234. The relevant parameters are listed in Table B2. The source area is assumed to be located at the inlet boundary (x=0) and the intervals are $20\ \mathrm{m} \le y \le 30\mathrm{m}$ and $20\ \mathrm{m} \le z \le 30\ \mathrm{m}$. The source function for exponential decay is used in this example. The user needs to check "Exponential Decay" on the source related parameter page, as shown in Figure B3, and further enter the "Source Decay Rate" and "Source Concentration" at the contamination source. In addition, select the "Transport Properties" of the decay rate parameter by checking "Decay occurs in both the dissolved phase and the adsorbed phase". The other parameters can be input according to Table B2. The simulation is then run to obtain the concentration profile with distance, as shown in Figure B4.

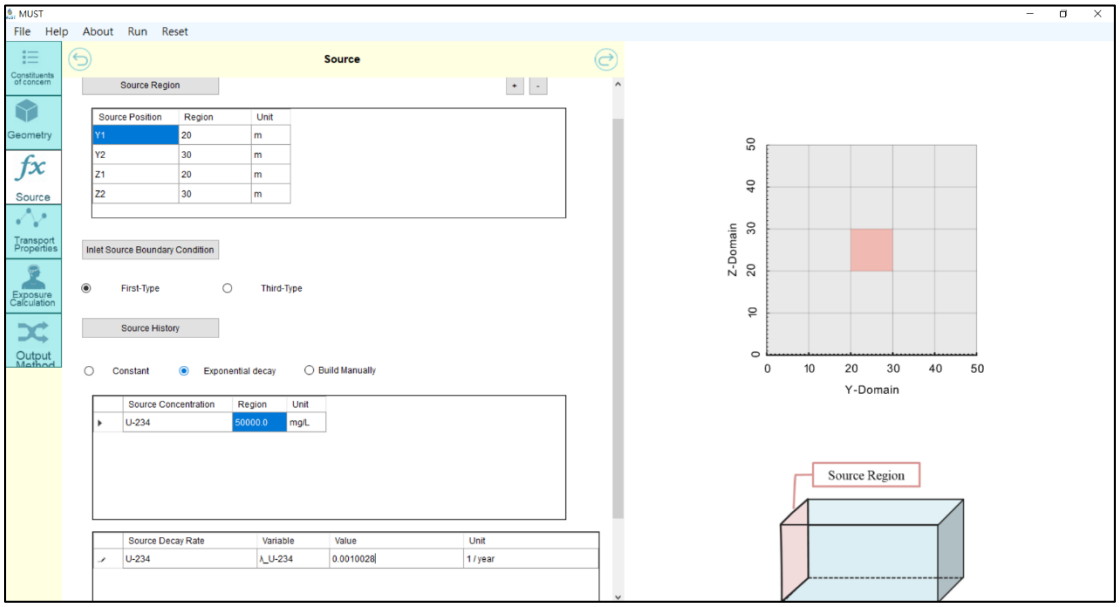


Figure B3. Selection of source functions.

Table B2. Parameters for Case 2 (Moranda et al., 2018).

Parameters	Value
Length, L [m]	100
Width, W [m]	50
Height, H [m]	50
Seepage velocity, v [m year^{-1}]	100
Longitudinal dispersion coefficient, D_x [m^2 year^{-1}]	4,000
Transverse dispersion coefficient, D_y [m^2 year^{-1}]	400
Vertical dispersion coefficient, D_z [m^2 year^{-1}]	400
Retardation factor, R_1 [-]	10,000
Decay rate constant, μ_1 [year^{-1}]	0.0000028
Source decay rate, λ_1 [year^{-1}]	0.0010028
Source concentration, $C_{1,0}$ [mg L^{-1}]	50,000

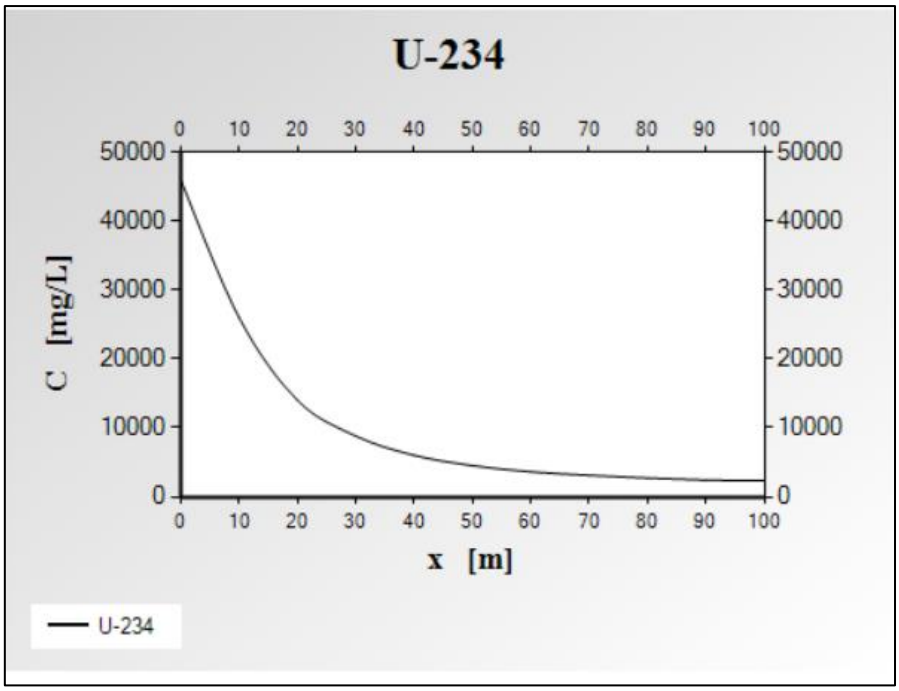


Figure B4. Concentration profile along the x-axis (y =25 m, z = 25 m) at 100 years.

Case 3. Single species transport: Effects of the dispersion coefficient

This simulation scenario is adapted from one of the illustrative examples used in 3DADE (Leij and Bradford, 1994). In this case, there is three-dimensional solute transport without considering decay and sorption. In this case, different longitudinal dispersion coefficients are used to investigate the effects of the dispersion on transport. The transport parameters for this example are summarized in Table 1. The source areas are located between $20\,\mathrm{m} \le y \le 40\mathrm{m}$ and $25\mathrm{m} \le z \le 35\,\mathrm{m}$ using the third-type boundary condition. Figure B5 shows a comparison between the spatial concentration profiles obtained using four different longitudinal dispersion coefficients (5, 10, 40, 100) along a straight line oriented in the *x* direction and going through the center of the rectangular source at 1 year. It can be seen that a larger dispersion coefficient may affect both the concentration level and mobility along the groundwater flow direction.

Table B3. Parameters for Case 3 (Leij and Bradford, 1994).

Parameters	Value
Length, L [m]	50
Width, W [m]	60
Height, H [m]	60
Seepage velocity, v [m year^{-1}]	20
Longitudinal dispersion coefficient, D_x [m^2 year^{-1}]	5, 10, 40, 100
Transverse dispersion coefficient, D_y [m^2 year^{-1}]	20
Vertical dispersion coefficient, D_z [m^2 year^{-1}]	10
Retardation factor, R_1 [-]	1
Decay rate constant, μ_1 [year^{-1}]	0
Source concentration, $C_{1,0}$ [mg L^{-1}]	1

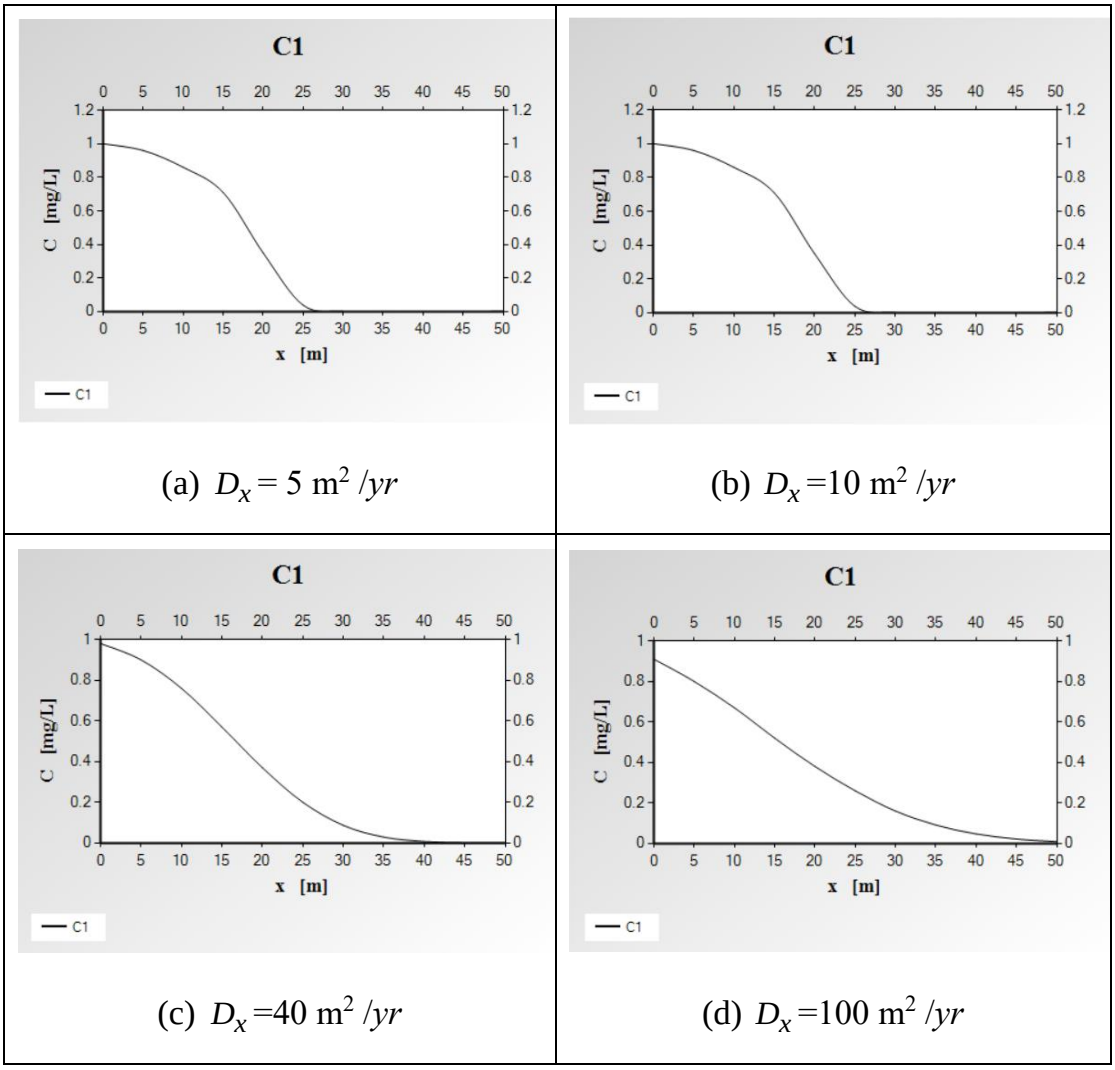


Figure B5. Numerical comparison of spatial concentration profiles obtained using four different longitudinal dispersion coefficients along a straight line oriented in the x direction at t = 1 year.

Case 4. Multispecies transport: Three nitrogen transformed products

This case describes the transport of nitrogen transformed products. This differs from the previous three cases is that this is a multispecies scenario with three contaminants, as shown in Figure B6. The reaction and transport parameters (please refer to van Genuchten, 1985) are listed in Table B4. Figure B7 shows the concentration profile of the three nitrogen transformed products, namely NH_4^+, NO_2^- and NO_3^-, along the direction of groundwater flow after 200 hours. It can be seen from the results that NH_4^+ was mainly released into the groundwater at the beginning while the concentrations of NO_2^- and NO_3^- started to increase and then to decrease with the decrease in the concentration of the parent species along the *x* direction, after a very short period of time.

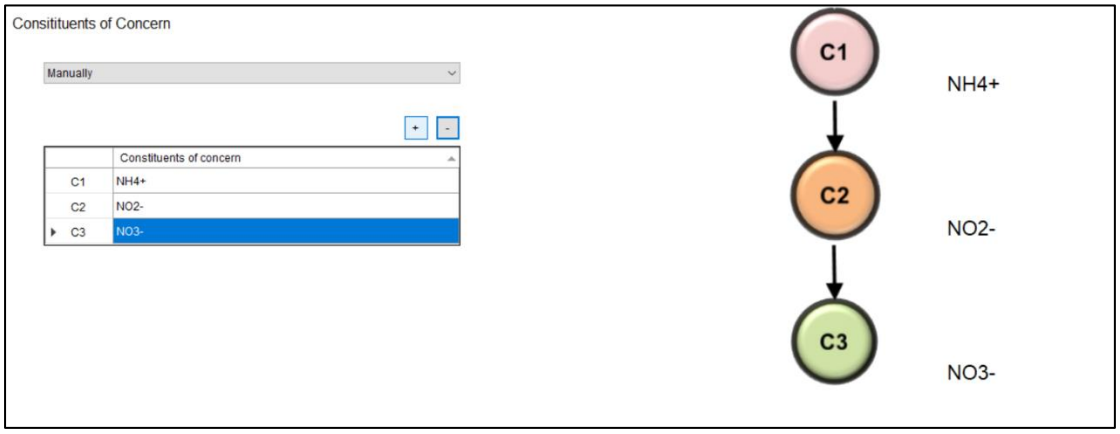


Figure B6. Adding multiple contaminants for the nitrogen transformed products on the interface.

Table B4. Parameters for Case 4 (van Genuchten, 1985)

Parameters	Value
Length, L [m]	200
Seepage velocity, v [cm h^{-1}]	1

Longitudinal dispersion coefficient, D_x [cm^2 h^{-1}]	0.18
Retardation factor, R_i [-]	
NH_4^+	2.0
NO_2^-	1.1
NO_3^-	1.0
Decay rate constant, μ_i [h^{-1}]	
NH_4^+	0.005
NO_2^-	0.1
NO_3^-	0.0
Source concentration, $C_{i,0}$ [mg L^{-1}]	
NH_4^+	1.0
NO_2^-	0.0
NO_3^-	0.0

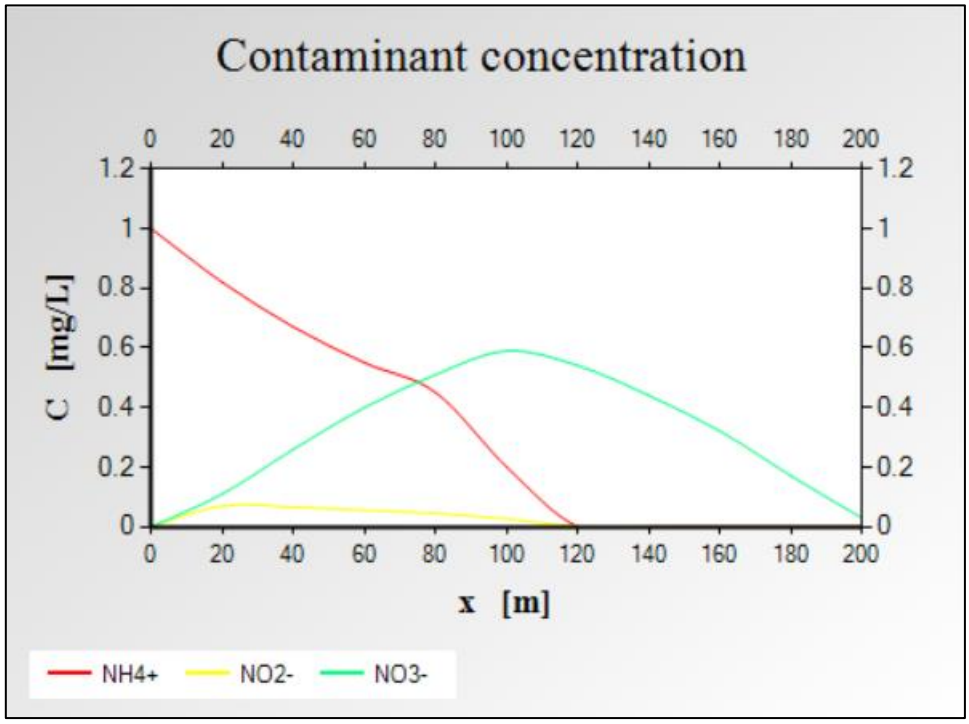


Figure B7. Concentration profile for the three nitrogen transformed products along the groundwater flow direction at 200 hours.

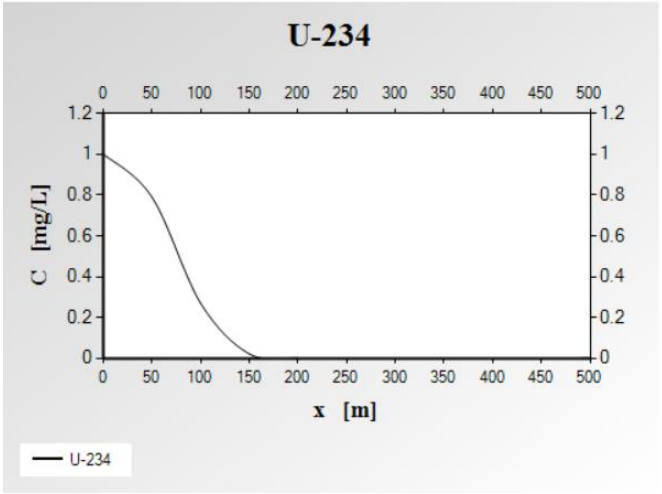
Case 5. Multispecies transport: Three radionuclides

This case simulates the transport of multiple radionuclides in a one-dimensional situation. The biggest difference from the radionuclide example in Case 2, is that this case considers both the initially released nuclide and the daughter products produced by decay reactions which results in a decay chain with three species in total. In this case, the initially released U-234 decays to produce the daughter species of Th-230 and Ra-226, as described in Sudicky et al. (2013). The parameters are listed in Table B5, and the yield coefficients are all set to 1. Figure B8 shows the simulation results for a simultaneous evaluation of the concentration profiles of all nuclides along the x direction after 10,000 years. The results show that there has been a significant decline in the first two radionuclides in the decay chain at a distance of 200 meters from the source. However, the third nuclide, Ra-226, has a smaller retardation factor and high mobility, so its distribution and distance of migration is large.

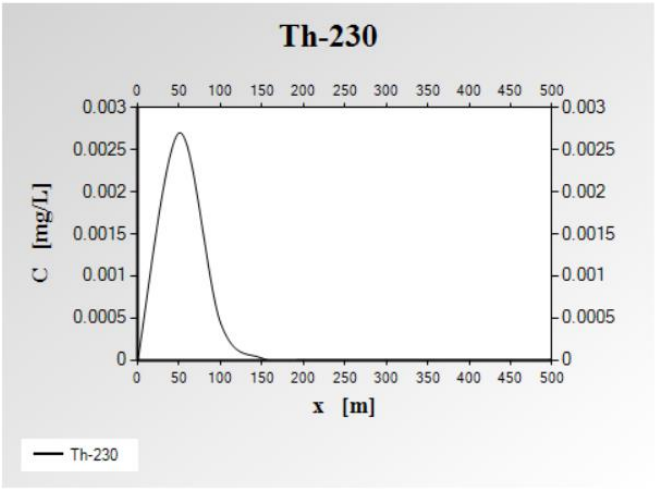
Table B5. Parameters for Case 5 (Sudicky et al., 2013).

Parameters	Value
Length, L [m]	500
Seepage velocity, v [m year^{-1}]	100
Longitudinal dispersion coefficient, D_x [m^2 year^{-1}]	1,000
Retardation factor, R_i [-]	
U^{234}	14,300
Th^{230}	50,000
Ra^{226}	500
Decay rate constant, μ_i [year^{-1}]	
U^{234}	0.00000283
Th^{230}	0.000009

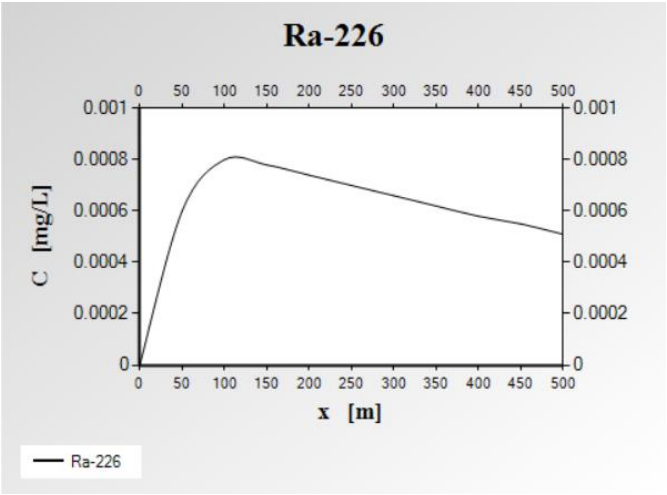
Ra^{226}	0.00000433
Source concentration, $C_{i,0}$ [mg L^{-1}]	
U^{234}	1.0
Th^{230}	0.0
Ra^{226}	0.0



(a)



(b)



(c)

Figure B8. Concentration profiles of the three radionuclides (a) U-234, (b) Th-230, (c) Ra-226 along the groundwater flow direction at 10,000 years.

Case 6. Multispecies transport: Four radionuclides

Case 6 also describes the transport of a multiple species decay chain with four radionuclides (Bauer et al., 2001). This case is also a one-dimensional scenario. The parameters are listed in Table B6, and the yield coefficients are all set to 1.The simulation results shown in Figure B9 help us to understand the concentration profiles of the original nuclide and the other products in the decay chain along the *x* direction after 3,000 days. In addition, we can further investigate the breakthrough curve of the concentration downstream from the source. Figure B10 shows the changes in the concentration with time at 5 m from the source. This can help us understand how long it will take for the radionuclide and its decay products to move to this location.

Table B6. Parameters for Case 6 (Bauer et al., 2001).

Parameters	Value
Length, L [m]	3,000
Seepage velocity, v [m day^{-1}]	1
Longitudinal dispersion coefficient, D_x [m^2 day^{-1}]	10
Retardation factor, R_i [-]	
C_1	5.3
C_2	1.9
C_3	1.2
C_4	1.3
Decay rate constant, μ_i [day^{-1}]	
C_1	0.0007
C_2	0.0005
C_3	0.00045
C_4	0.00038
Source concentration, $C_{i,0}$ [mg L^{-1}]	
C_1	100.0
C_2	0.0

C_3	0.0
C_4	0.0

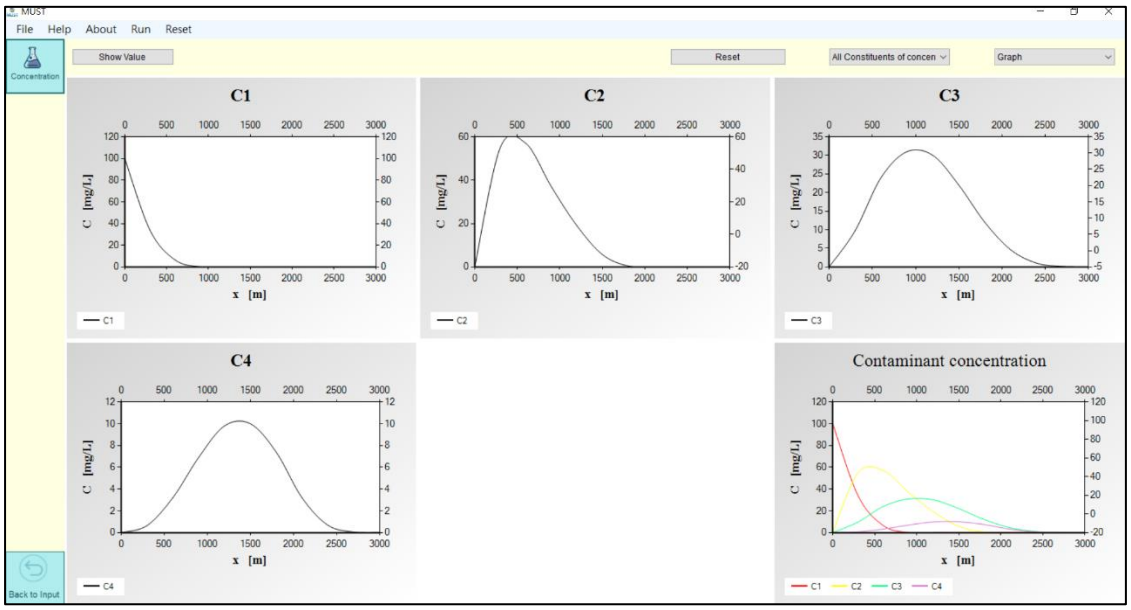


Figure B9. Concentration profiles of the four radionuclides along the *x* direction at 3,000 days; the simulation results for all nuclides appear in the right-hand corner.

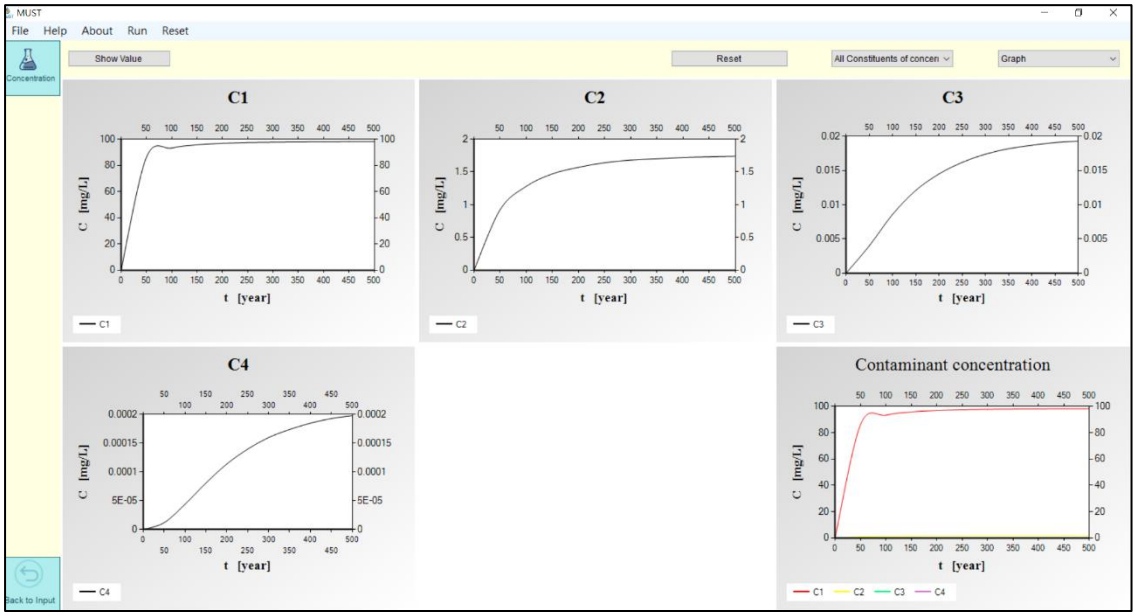


Figure B10. Changes in concentration with time of the four radionuclides at 5 m from the source.

Case 7. Multispecies transport: Five chlorinated solvents

Case 7 is the same as the example of two-dimensional transport discussed in chapter 3 which refers to a chlorinated solvent contaminated site at Cape Canaveral Air Station in Florida. This illustrative example was designed to reproduce the movement of the plumes from the site over the previous 30. The contaminants involve a five-species chlorinated solvent decay chain: tetrachloroethylene (PCE) $\rightarrow$ trichloroethylene (TCE) $\rightarrow$ dichloroethylene (DCE) $\rightarrow$ vinyl chloride (VC) $\rightarrow$ ethylene (ETH). The simulation conditions and transport parameters for this case are summarized in Table B7 from the BIOCHLOR model (Aziz et al., 2000). The source area is assumed to be located between $20\ \mathrm{m} \le y \le 30\mathrm{m}$.

Figure B11 visualizes the two-dimensional spatial concentration contours of the five chlorinated solvents on the *x*-*y* plane at five different times (t = 4, 8, 12, 16, 20 years). The results show that the daughter products, VC and ETH, initially have lower concentrations, but their concentrations increase as the plume size becomes larger.

To investigate the effect of the retardation factor values on the multiple contaminant transport, the performance of the previously developed model is assessed using the same retardation factor for all species. Figures B12a and B12b show the resultant spatial concentration profiles of the five species along x direction at 5 years produced using individual retardation factors and approximately the same retardation factor (median value).

According to the results, the larger retardation value of PCE ($R_{\mathrm{PCE}} = 7.13$) causes the plume to become smaller than for the other species. However, for VC ($R_{\mathrm{VC}} = 1.43$), whose retardation factor value is smaller, the plume size is larger because of its higher mobility and its daughter product, ETH, would also have a large area as a result.

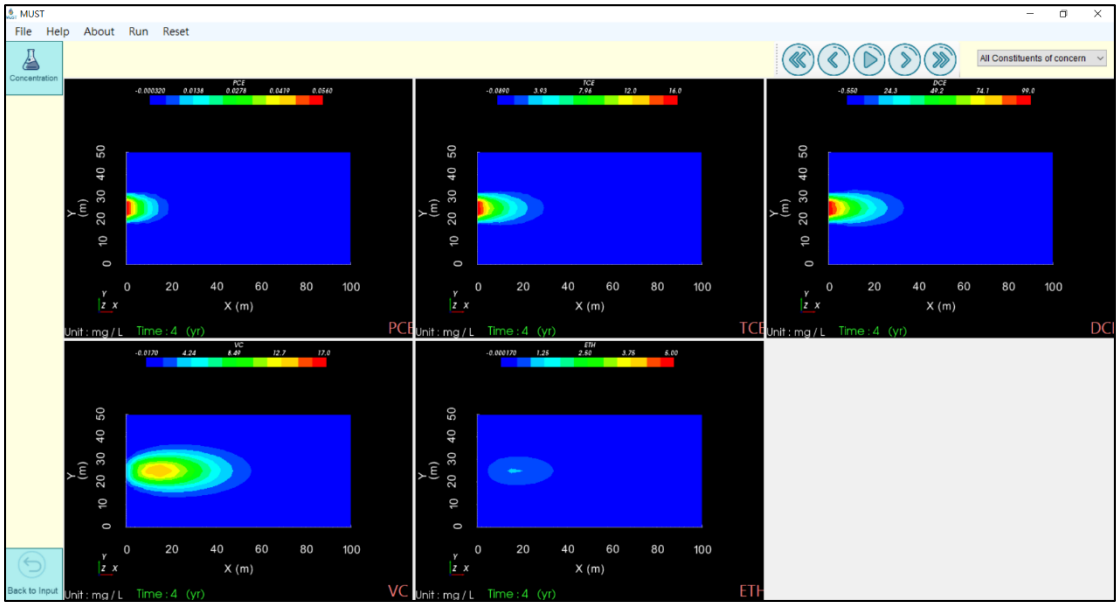
For an assessment of the performance of the previous model using the same

retardation factor, the simulation results are used to find whether there is an overestimation or underestimation of the concentration for different species. The software simulation comparisons demonstrate the difference it makes when incorrect parameters are considered for simulating multispecies plume migration of degradable contaminants in the field application.

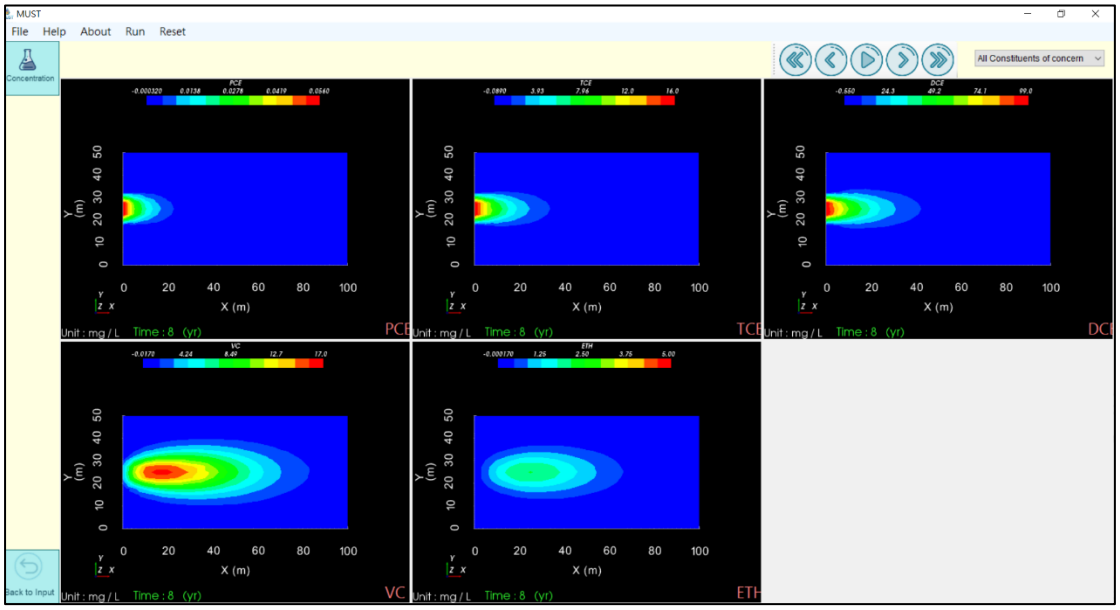
Table B7. Parameters for Case 7 from BIOCHLOR (Aziz et al., 2000).

Parameters	Value
Length, L [m]	100
Width, W [m]	50
Seepage velocity, v [m year^{-1}]	10
Longitudinal dispersion coefficient, D_x [m^2 year^{-1}]	100
Horizontal transverse dispersion coefficients, D_y [m^2 year^{-1}]	10
Retardation factor, R_i [-]	
PCE	7.13
TCE	2.87
DCE	2.8
VC	1.43
ETH	5.35
Decay rate constant, μ_i [year^{-1}]	
PCE	2.0
TCE	1.0
DCE	0.7
VC	0.4
ETH	0.0
Source concentration, $C_{i,0}$ [mg L^{-1}]	
PCE	0.056
TCE	15.8
DCE	98.5
VC	3.08

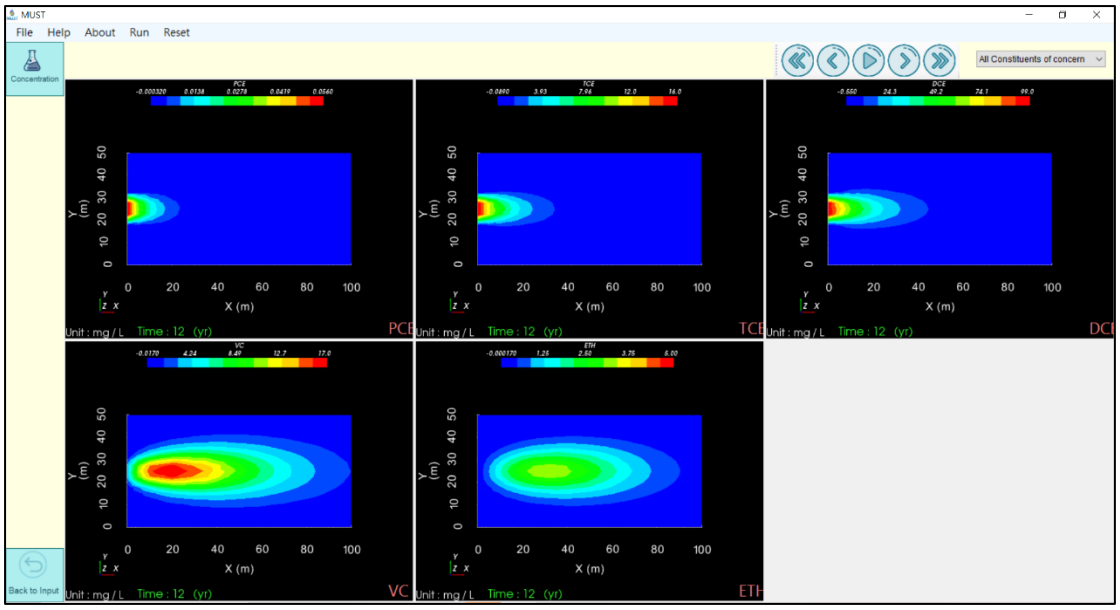
ETH	0.03
Yield coefficient, $Y_{i-1\rightarrow i}$ [-]	
$Y_{\mathrm{PCE}\rightarrow\mathrm{TCE}}$	0.79
$Y_{\mathrm{TCE}\rightarrow\mathrm{DCE}}$	0.74
$Y_{\mathrm{DCE}\rightarrow\mathrm{VC}}$	0.64
$Y_{\mathrm{VC}\rightarrow\mathrm{ETH}}$	0.45



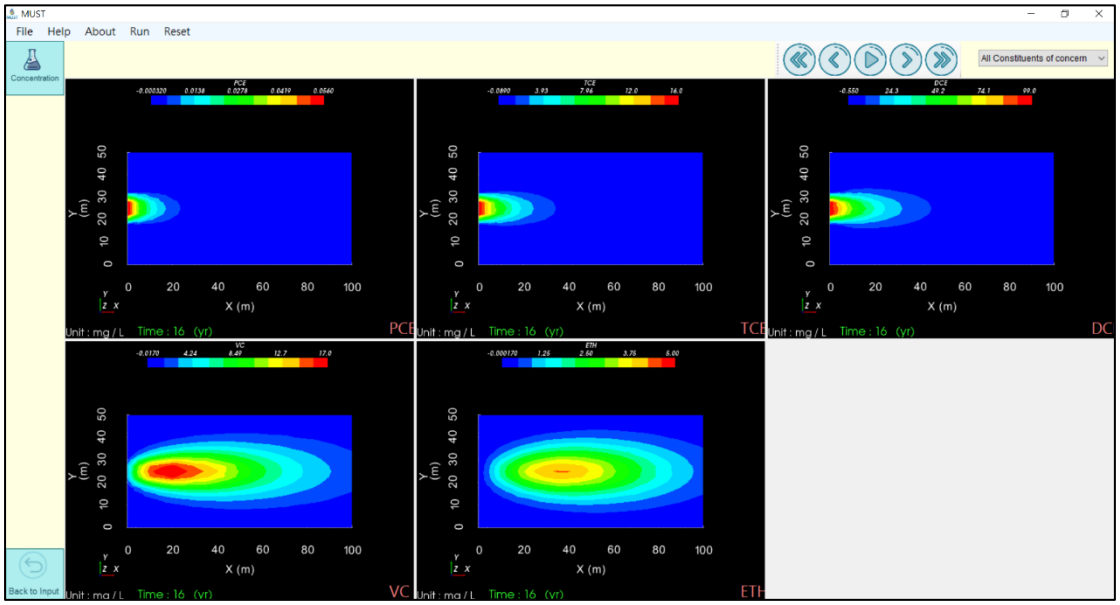
(a) 4 years



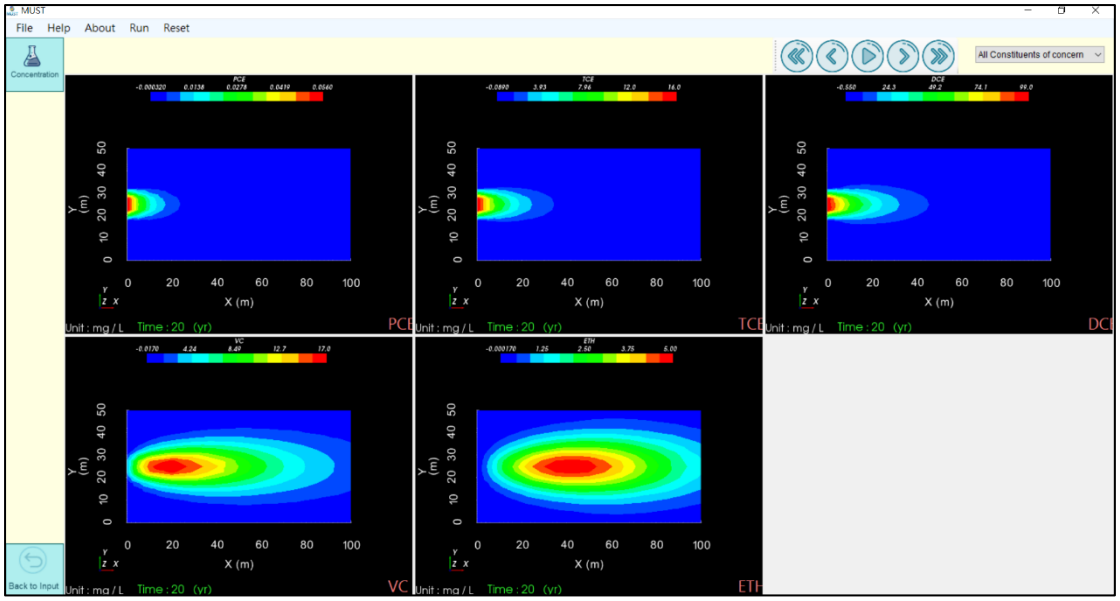
(b) 8 years



(c) 12 years



(d) 16 years



(e) 20 years

Figure B11. Animation of the plume migration for chlorinated solvents at: (a) 4 years (b) 8 years (c) 12 years (d) 16 years (e) 20 years.

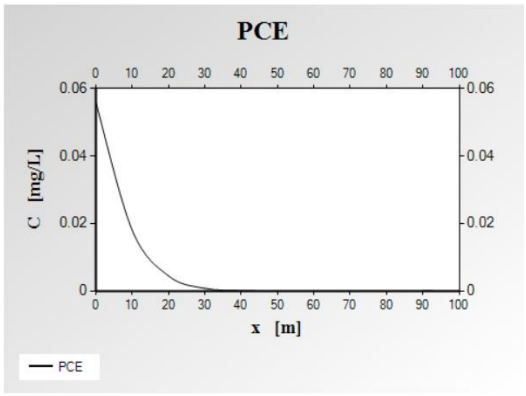
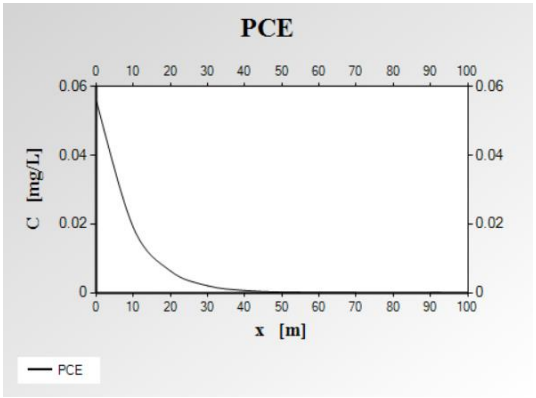
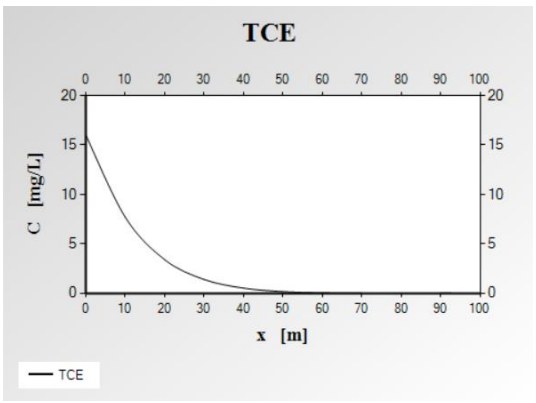
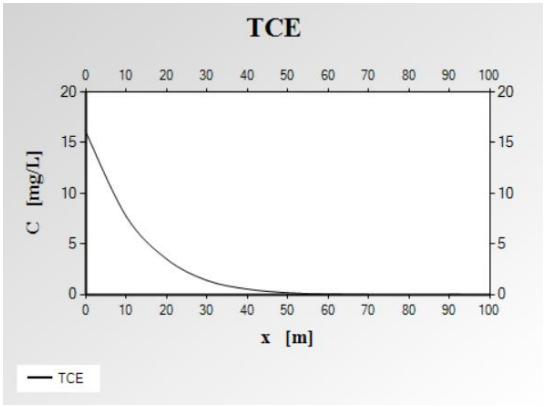
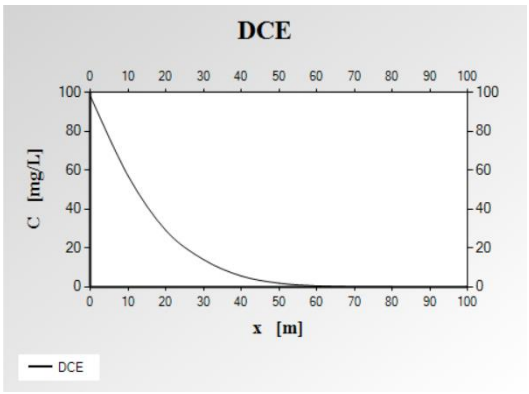
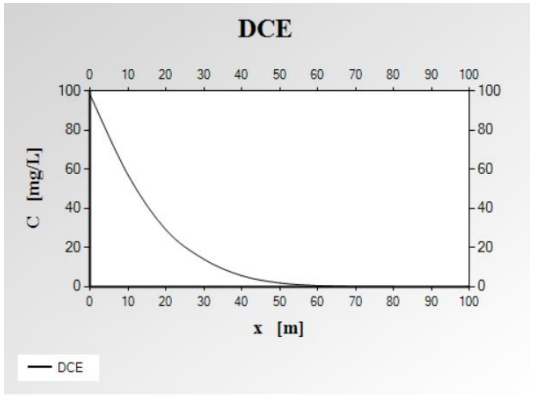
Individual retardation factor used for all species	Similar retardation factor of 2.8 used for all species
	
	
	

Figure B12a. Simulation results showing the concentration profiles of PCE, TCE and DCE along the *x* direction at 5 years obtained using different and similar retardation factors.

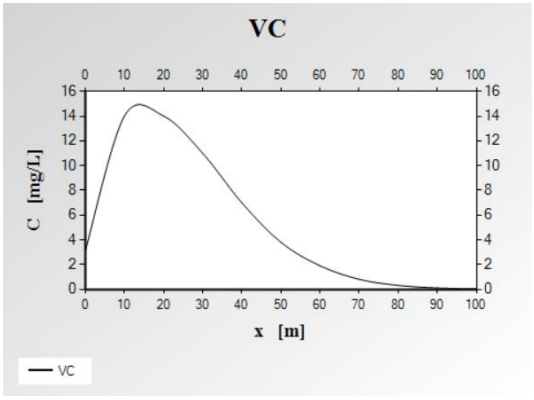
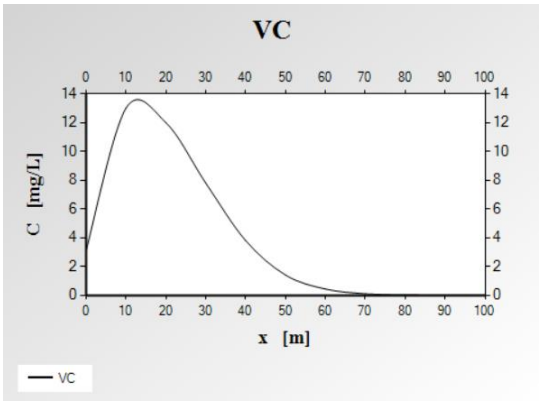
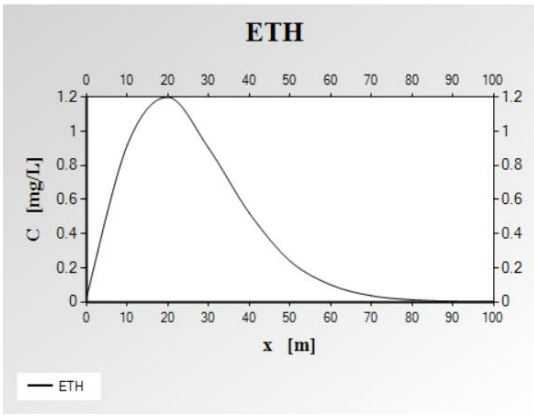
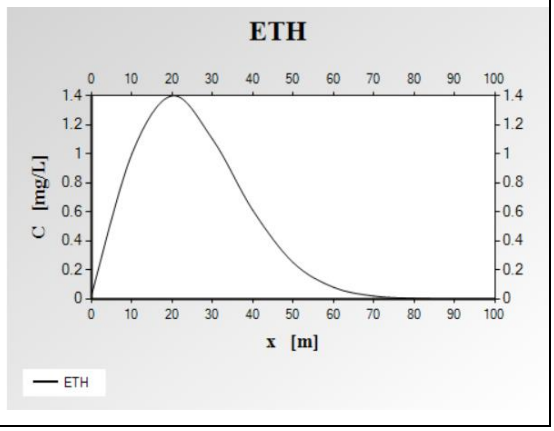
Individual retardation factors are used for all species	Similar retardation factor of 2.8 used for all species
	
	

Figure B12b. Simulation results showing the concentration profile of VC and ETH along the *x* direction at 5 years obtained using different and similar retardation factors.

Case 8. Multispecies transport: Source geometry and numbers

This case is mainly a continuation of the example of multispecies transport of chlorinated solvents in Case 7 and simulates the situation where multiple contaminated sources exist at the same time. The relevant reaction and transport parameters are the same as those in Case 7. The biggest difference is in the numbers, area and source concentration of the sources. In this case, the user must add two more sources on the screen related to the source parameters, as shown in Figure B13. These three sources are in the regions of $20\ \text{m} \le y \le 30\text{m}$, $0\ \text{m} \le y \le 10\text{m}$, $30\ \text{m} \le y \le 35\text{m}$,. The different sources are highlighted in different colors on the right side for ease of visualization. Table B8 lists the concentration values of each contaminant at different sources. These values need to be input individually at the Source History screen, as shown in Figure B14. Figure B15 shows the simulated plume migration of five chlorinated solvents after 20 years after setting the values of the three contaminants released at the source. The simulation results further our understanding of how the difference in the source region will affect the plume size. The source concentration values in different regions will also affect the distribution and position of the hot spot.

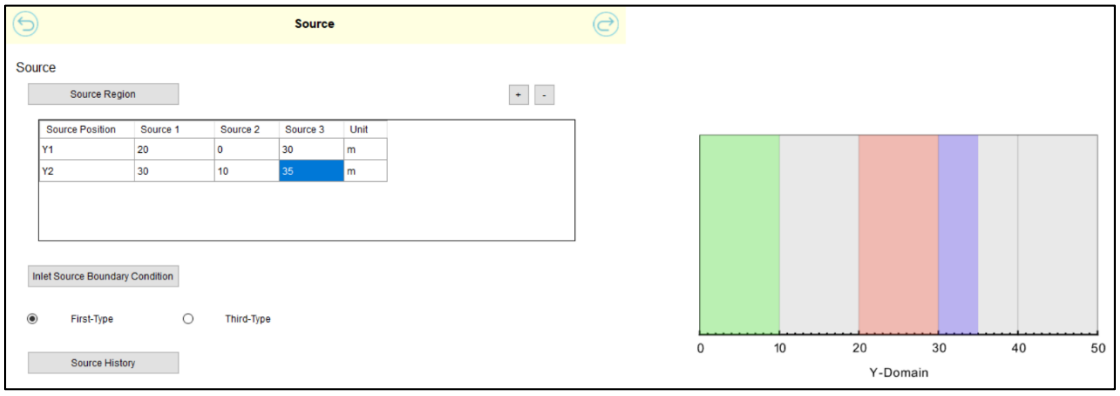


Figure B13. Building multi-sources on the interface.

Table B8. Source concentrations of five chlorinated solvents for three sources.

Source concentration (mg/L)	Source 1	Source 2	Source 3
PCE	0.056	0.5	0.05
TCE	15.8	10	1
DCE	98.5	100	20
VC	3.08	10	5
ETH	0.03	1	2

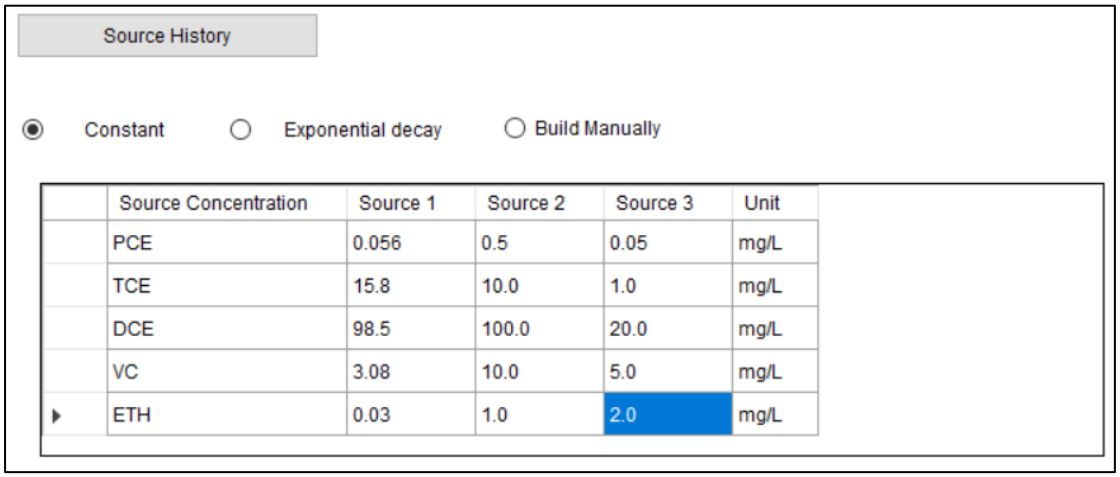


Figure B14. Input the source concentrations for the different contaminants on the user interface.

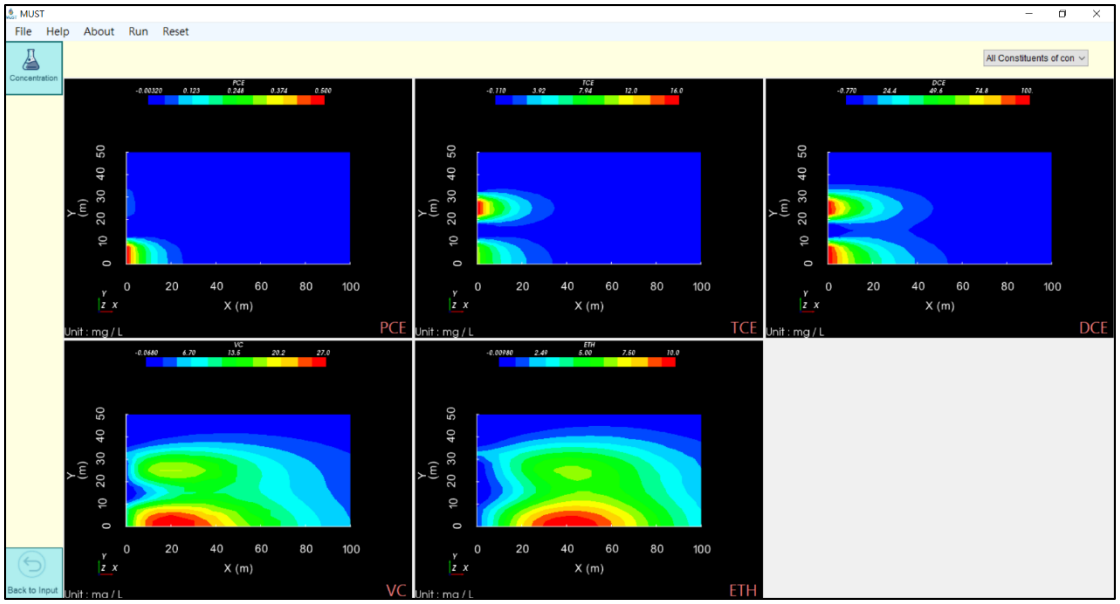


Figure B15. The plume migration of five chlorinated solvents at 20 years with three release sources set.

Case 9. Multispecies transport: Application of time-dependent source functions

In the Case 9 model, the source concentrations change over time. This application refers to the third case in the REMChlor model manual (Falta, 2007). This example considers two contaminants, with initial exposure to the chlorinated solvent 1,1,1-trichloroethane (1,1,1-TCA) which further produces the degradation product of 1,1-dichloroethane (1,1-DCA). In the REMChlor manual (Falta, 2007), the aim is to assess the remediation at the site coupled with both source and plume remediation. In the REMChlor model, the source function describes the relative relationships by combining the relational expressions including the source mass, concentration and empirical parameters. The empirical parameter used by REMChlor in this case has a value of 2. However, in the MUSt software, it is only necessary to input the source concentrations at different times, which is simpler and more useful than the previous models.

The steps are as follows. First, select the two-dimensional scenario, and then click the third-type inlet boundary condition and check the source function for "Build Manually." The source concentration is defined at different times according to the same relationship defined by the formula in the REMChlor manual. Figure B16 shows the table for entering the concentration values after selecting "Build Manually" on the user interface. The input reaction and transport parameters related to the model are listed in Table B10.

The software can describe changes in the trend of the concentration at the contaminated source (x=0) by inputting the source concentrations at different times, as shown in Figure B17. It can also simulate the concentration breakthrough curves at different downstream observation points, as shown in Figure B18. In addition, the concentration profiles at different distances at different times can also be simulated, as shown in Figure B19. The application for this case illustrates the practicality of this

software model for simulating the observed changes in concentration at contaminated sites.

Table B9. Source concentration of two chlorinated solvents at different times (mg/L).

Time/species	0	1yr	2yr	3yr	4yr	5yr	6yr	7yr	8yr	9yr
TCA	2	1.93	1.89	1.85	1.79	1.74	1.7	1.67	1.64	1.62
DCA	0	0	0	0	0	0	0	0	0	0

Figure B16. Table for inputting source concentrations after selecting "Build Manually".

Table B10. Parameters for case 9 from REMChlor (Falta, 2007).

Parameters	Value
Length, L [m]	200
Width, W [m]	100
Seepage velocity, v [m year^{-1}]	60
Longitudinal dispersion coefficient, D_x [m^2 year^{-1}]	50
Horizontal transverse dispersion coefficients, D_y [m^2 year^{-1}]	5
Retardation factor, R_i [-]	
1,1,1-TCA	5.56
1,1-DCA	1.91
Decay rate constant, μ_i [year^{-1}]	
1,1,1-TCA	0.8
1,1-DCA	0.2
Source concentration, $C_{i,0}$ [mg L^{-1}]	

1,1,1-TCA	2
1,1-DCA	0
Yield coefficient, $Y_{i-1\rightarrow i}$ [-]	
$Y_{TCA\rightarrow DCA}$	0.74

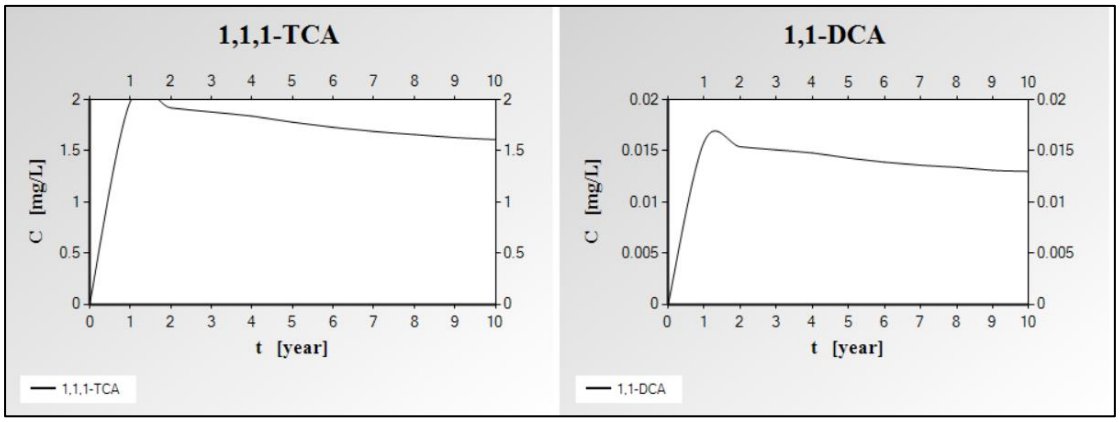
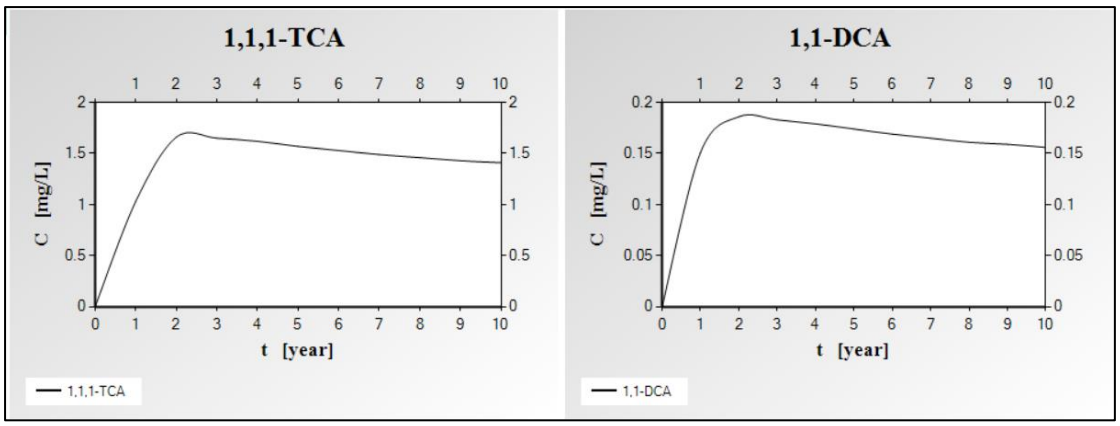
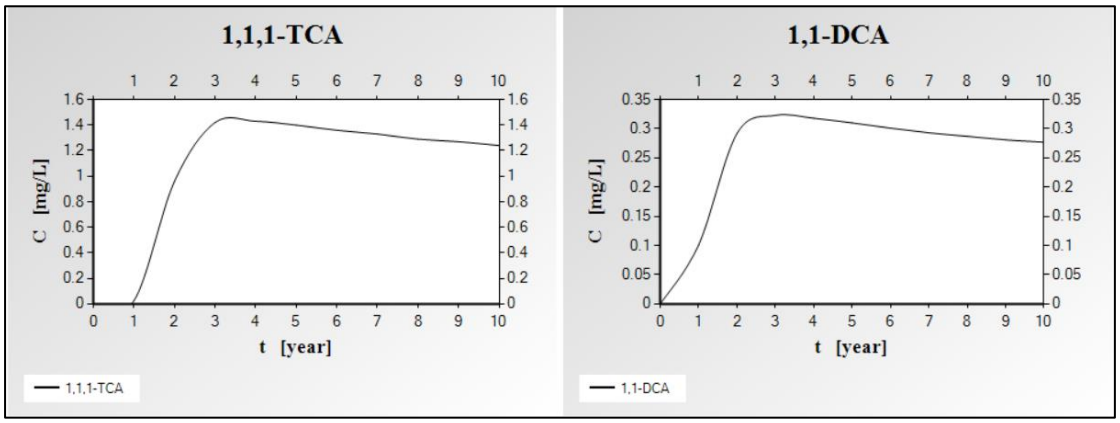


Figure B17. Concentration breakthrough curves at the source（x=0, y=50 m).



(a) x=10 m



(b) x=20 m

Figure B18. Concentration breakthrough curves at different downgradient observation points (a) x= 10 m; (b) x=20 m.

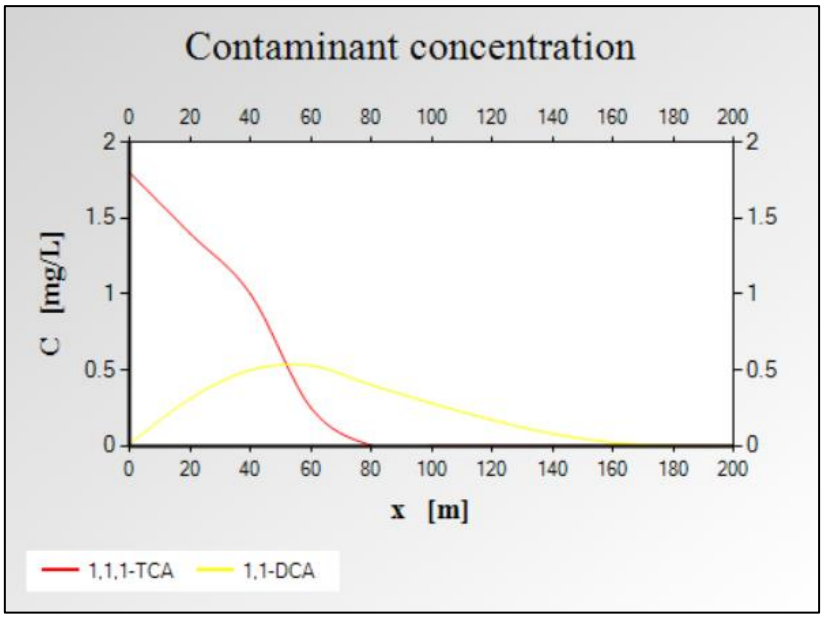


Figure B19. Concentration profile of two chlorinated solvents along x direction at 5 years.