Fate and contaminant transport model-driven probabilistic human health risk assessment of DNAPL-contaminated site

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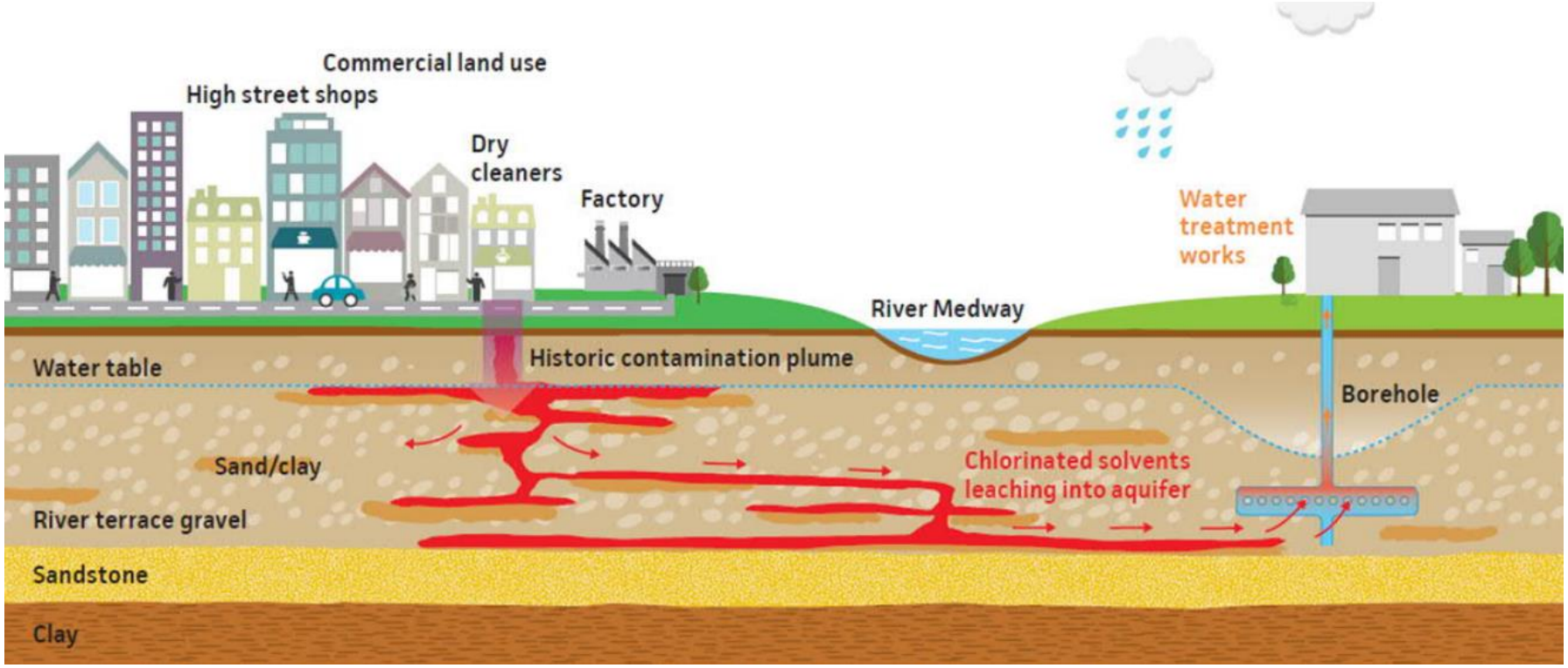
Outline

- Introduction
- Methodology
- Discussion and results
- Conclusions

Groundwater contaminants

- There are many contaminated sites worldwide, and the contamination of the subsurface environment pose threats to human health.

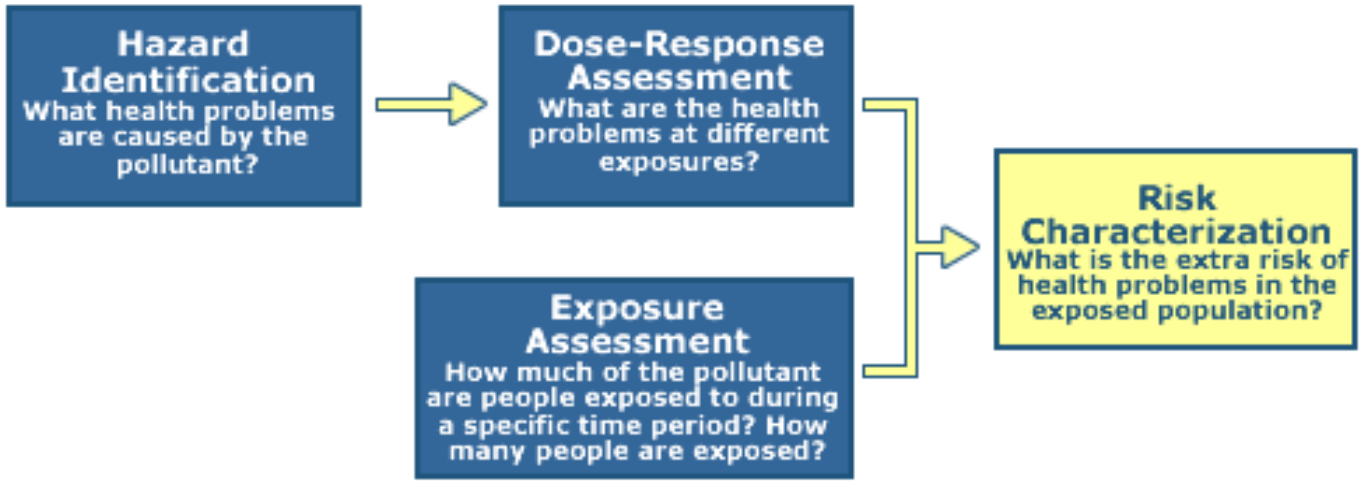
- Chlorinated solvents like tetrachloroethene (PCE) and trichloroethene (TCE) are common contaminants in groundwater that cause different kinds of cancer.



Human health risk assessment (HHRA)

- HHRA is the process to estimate the probability of adverse health effects in humans who may be exposed to chemicals in contaminated environmental media.

- HHRA can be the reference of the remedial actions, also can help governments to deliver technical knowledge to the general public.



Previous research

- 5th, 95th, median, and average value of health risks were calculated considering uncertainty in contaminant concentration only, while a single point value of exposure model parameters (IR, BW, ED, EF) was adopted in risk assessment framework (Liu et al. 2019).

- In some of the studies, point estimate approach for exposure model parameters was considered assuming direct oral ingestion exposure scenario only, neglecting the effect of uncertain exposure model parameters on risk indexes (Barros et al. 2016; Libera et al. 2019; Bai et al. 2019; Qiao et al. 2019).

Objective

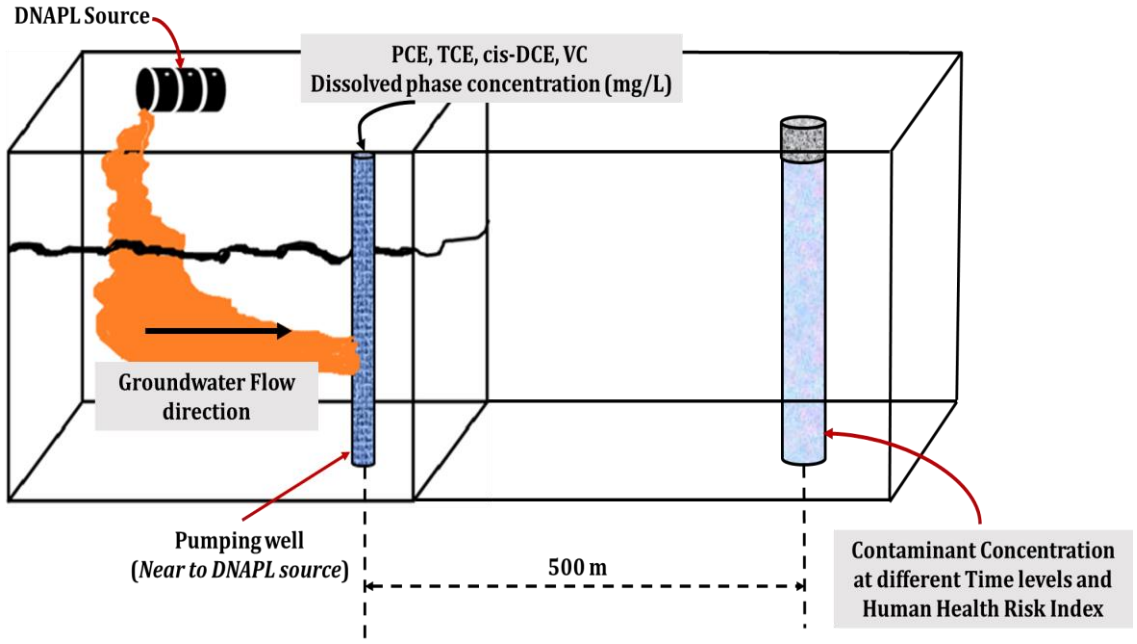
- Implement a probabilistic, contaminant transport model-driven human health risk assessment for a DNAPL-contaminated site:
  - To investigate the impact of longitudinal dispersivity on concentration.
  - To conduct risk assessment for the children and adults.
  - To compute non-carcinogenic and carcinogenic risk indexes for skin dermal contact and direct oral ingestion.
  - To assess the relative significance parameters on the overall uncertainty in risk estimates.

Contaminant transport model

- Governing equations representing transport and transformation can be described as:

$$\theta R_1 \frac{\partial C_1}{\partial t} = -\nabla \cdot (qC_1) + \nabla \cdot (\theta D \nabla C_1) - k_1 \theta C_1 + s(x,t) \qquad \text{for PCE}$$

$$\theta R_i \frac{\partial C_i}{\partial t} = -\nabla \cdot (qC_i) + \nabla \cdot (\theta D \nabla C_i) - k_i \theta C_i + y_i k_{i-1} \theta C_{i-1} \qquad \text{i=2 for TCE; i=3 for cis-DCE; i=4 for VC}$$



Where:
C_i: contaminant concentration (mg/L)
θ: porosity
q: Darcy velocity (m/day)
D: dispersion coefficient (m^2/day) $D = \alpha_L \frac{q}{\theta}$
k_i: first-order decay rate (day^{-1})
y_i: yield coefficient ($g\ g^{-1}$)
R_i: retardation factor (equilibrium sorption)

Exposure and dose-response assessment of this study

- Consider two exposure pathways:
  - Direct oral ingestion of groundwater as drinking water
  - Dermal contact through bathing

- Exposure dose (average daily dose) is calculated as:
  - Oral ingestion: $ADD_i = C_i \times \frac{IR \times EF \times ED}{BW \times AT}$
  - Dermal contact: $ADD_i = (DA_{event})_i \times \frac{SA \times EV \times EF \times ED}{BW \times AT}$

Where:
ADD_i: average daily dose (mg/kg-day)
C_i: contaminant concentration (mg/L)
$(DA_{event})_i$: absorbed dose taken in single event (mg/cm^2-event)
IR: water ingestion rate (L/day)
SA: skin surface area (cm^2)
EV: event frequency (events/day)
EF: exposure frequency (days/year)
ED: exposure duration (years)
BW: body weight (kg); AT: average time (days)
RfD: reference dose; SF: slope factor

- Non-carcinogenic and carcinogenic risk indexes are calculated as:
  - Non-carcinogenic: $R_i = \left(\frac{ADD}{RfD}\right)_i$
  - Carcinogenic: $R_i = ADD_i \times SF_i$

Approach of this study

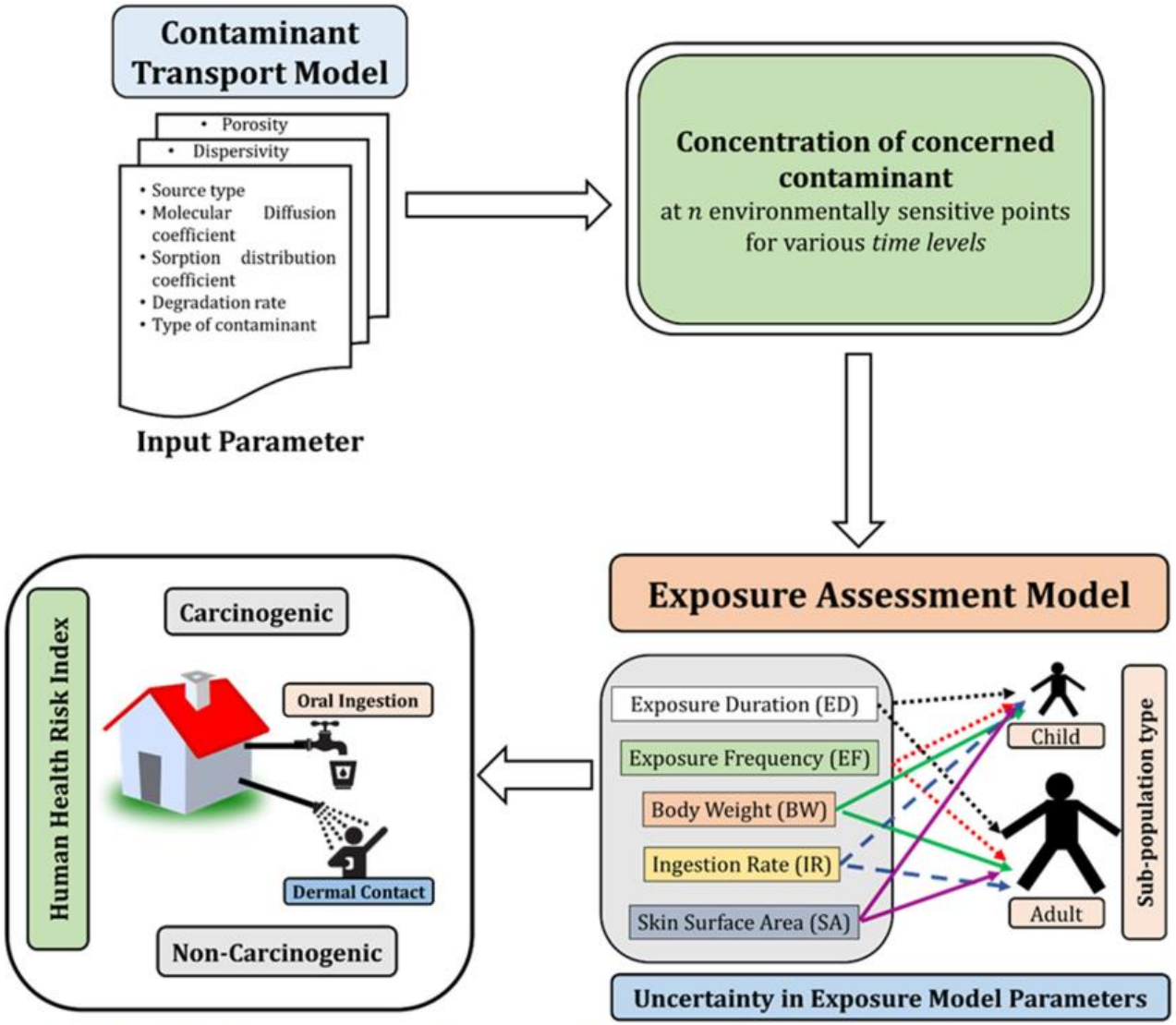


Fig. 1 Framework of fate and contaminant transport model-driven probabilistic human health risk assessment approach

Input parameters

Table 1 Input parameters used in the fate and contaminant transport model

Parameter		Value	Reference
Length (L)		500 m	Assumed
Porosity (θ)		0.30	Henri et al. (2015)
Darcy velocity (q)		0.20 m/day	Assumed
Contaminant source duration (t_{pulse})		1 year (365 days)	Assumed
Simulation time ($t_{simulation}$)		50 years (18,250 days)	Assumed
Longitudinal dispersivity (α_L) - normal distribution[#]		25, 5.0[2]	Assumed based on the observations (Gelhar et al. 1992; Schulze-Makuch 2005)
First-order decay rate constant (k_i)	k_{PCE}	0.0038 day^{-1}	Pivetz et al. (2014)
	k_{TCE}	0.0041 day^{-1}	
	$k_{cis-DCE}$	0.0020 day^{-1}	
	k_{VC}	0.0099 day^{-1}	
Yield coefficient for one to another component	$y_{PCE \rightarrow TCE}$	0.79 (g g^{-1})	Henri et al. (2016)
	$y_{TCE \rightarrow (cis-DCE)}$	0.74 (g g^{-1})	
	$y_{(cis-DCE) \rightarrow VC}$	0.64 (g g^{-1})	
Retardation factor (R_i)	R_{PCE}	7.1	Aziz e
	R_{TCE}	2.9	
	$R_{cis-DCE}$	2.8	
	R_{VC}	1.4	
Concentration at source up to t_{pulse} time	PCE	0.056 (mg/L)	Aziz e
	TCE	15.8 (mg/L)	
	Cis-DCE	98.5 (mg/L)	
	VC	3.08 (mg/L)	

[#] (Mean, standard deviation[2]); longitudinal dispersivity (units – meter)

Table 2 Risk assessment model parameters

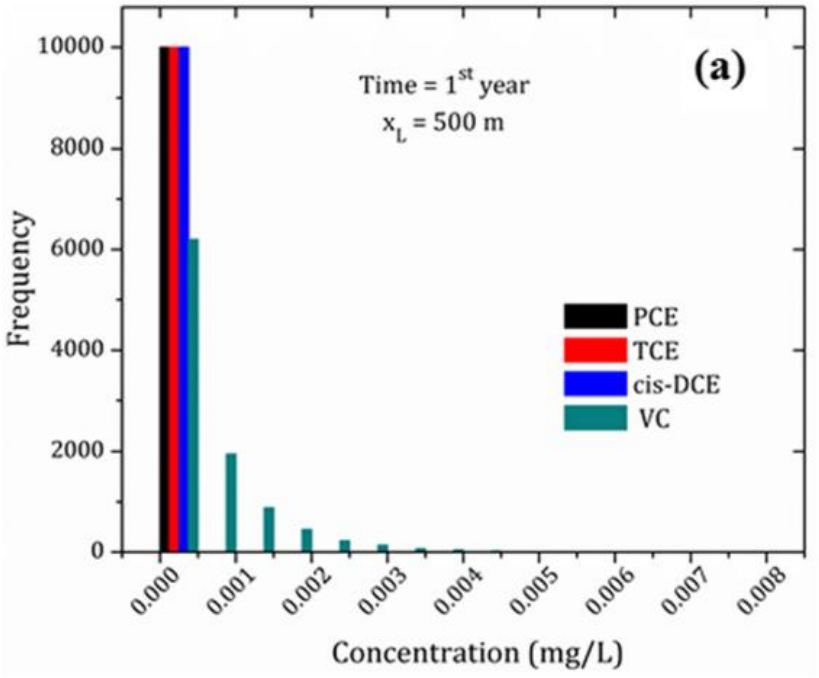
Parameter	Sensitive sub-population	Distribution type	Value*	Reference
Bodyweight (BW), kg	Children	Normal	16.67, 5.987[2]	Kumar and Xagoraraki (2010)
	Adults	Normal	70, 14[2]	
Ingestion rate, IR (L/day)	Children	Normal	1.25 ± 0.57	Fallahzadeh et al. (2018)
	Adults	Normal	1.95 ± 0.64	
Exposure duration, ED (years)	Children	Uniform	0, 5	USEPA (1997); Rajasekhar et al. (2018)
	Adults	Uniform	0, 50	
Exposure frequency, EF (days/year)	Children and adults	Triangular	Min: 180 Mode: 345 Max: 365	Fallahzadeh et al. (2018)
Skin surface area, SA (cm^2)	Children	Log-normal	5838, 920	USEPA (1997); Rajasekhar et al. (2018)
	Adults	Log-normal	19,771, 3373	
Single contact event duration, t_{event} (hour)	Children	Fixed value	0.33	Zhang et al. (2019); USEPA (2004)
	Adults	Fixed value	0.25	
Event frequency, EV	Both sub-population	Fixed value	1	Zhang et al. (2019); USEPA (2004)

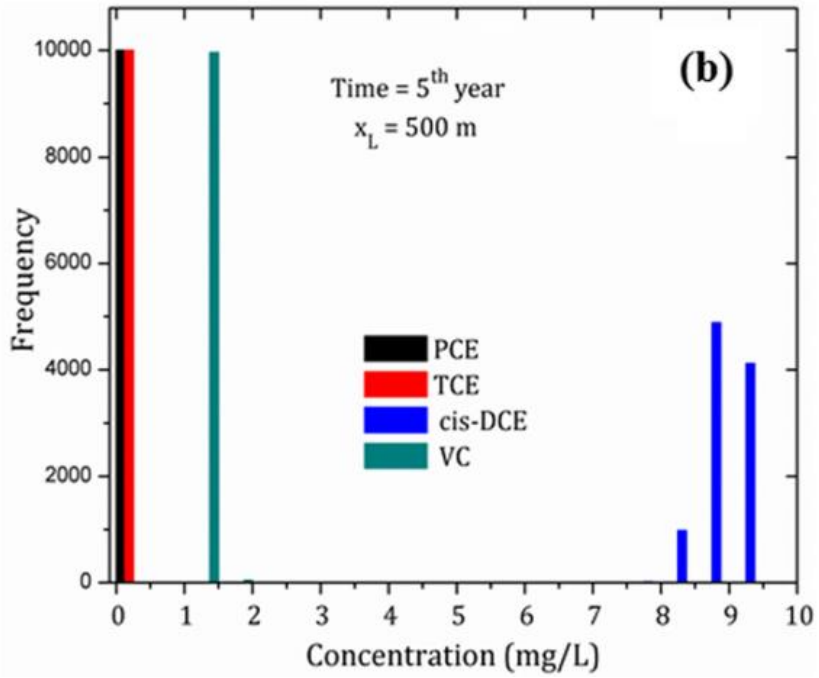
*Mean, standard deviation[2]

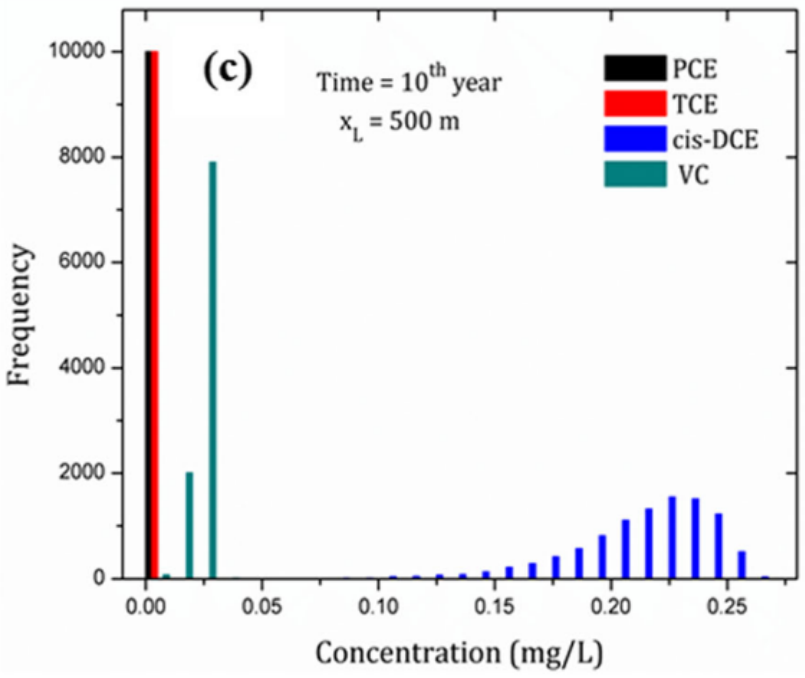
Longitudinal dispersivity effect

- Different compound has different transport behavior.

- Concentration decrease from 5th to 10th year represent the dilution of source concentration by dispersion processes with time.





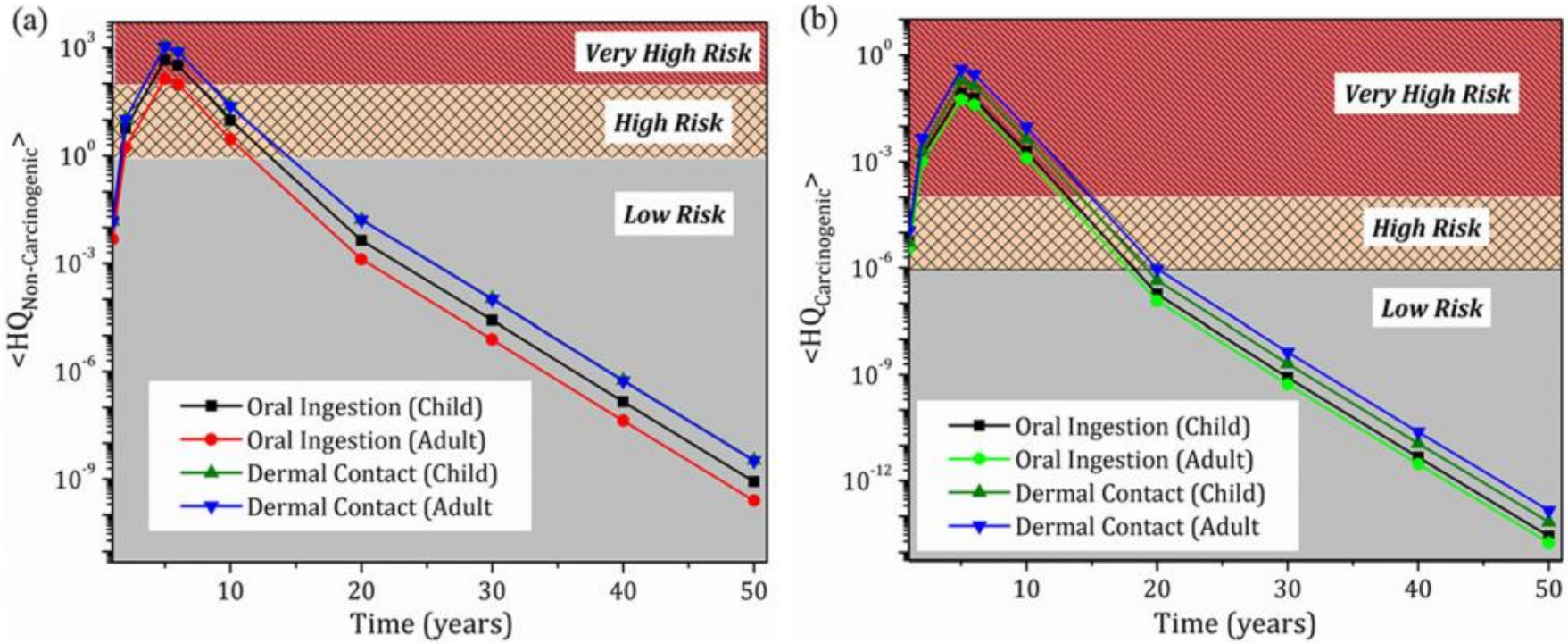


Total risk indexes

- Total risk of carcinogenic and non-carcinogenic is calculated as:

$$R_{\text{total}} = \Sigma R_i$$

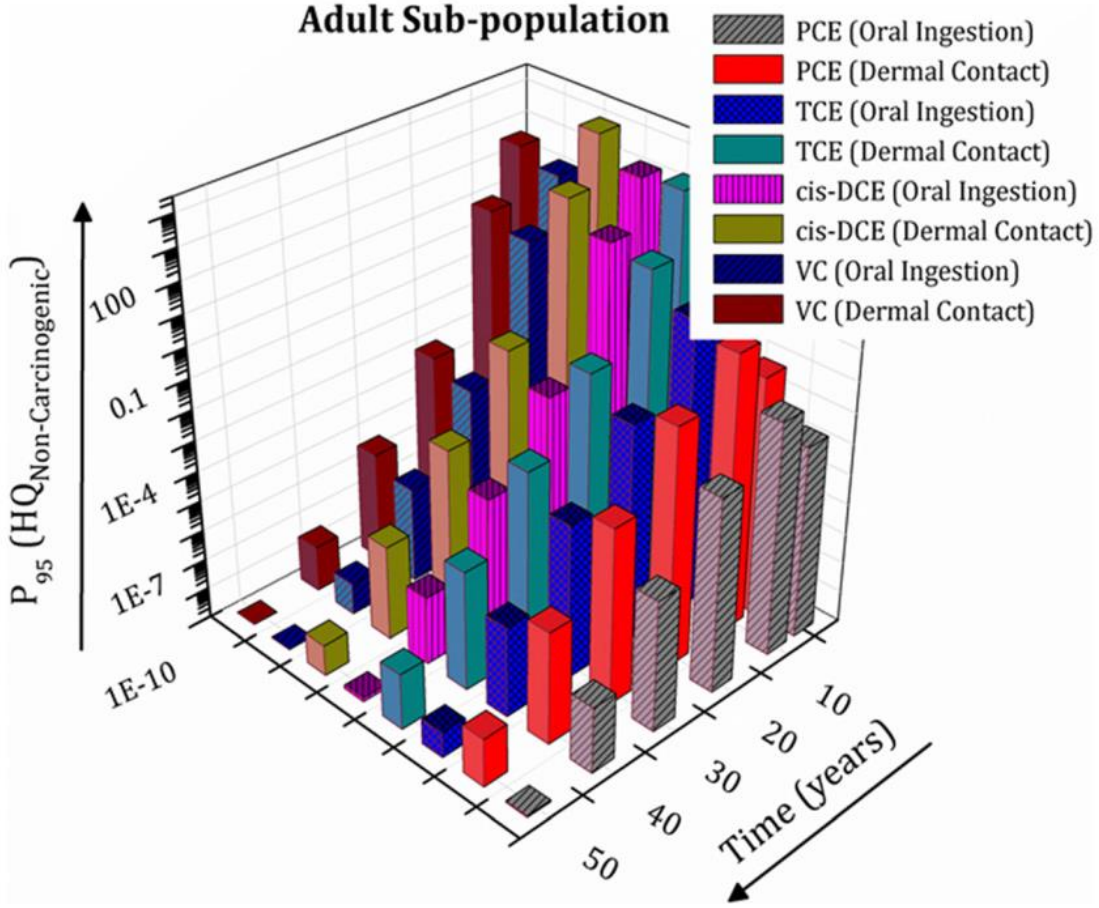
- These conditions represent a potential risk to human health:
  - Non-carcinogenic: $R \geq 1$
  - Carcinogenic: $R \geq 10^{-6}$

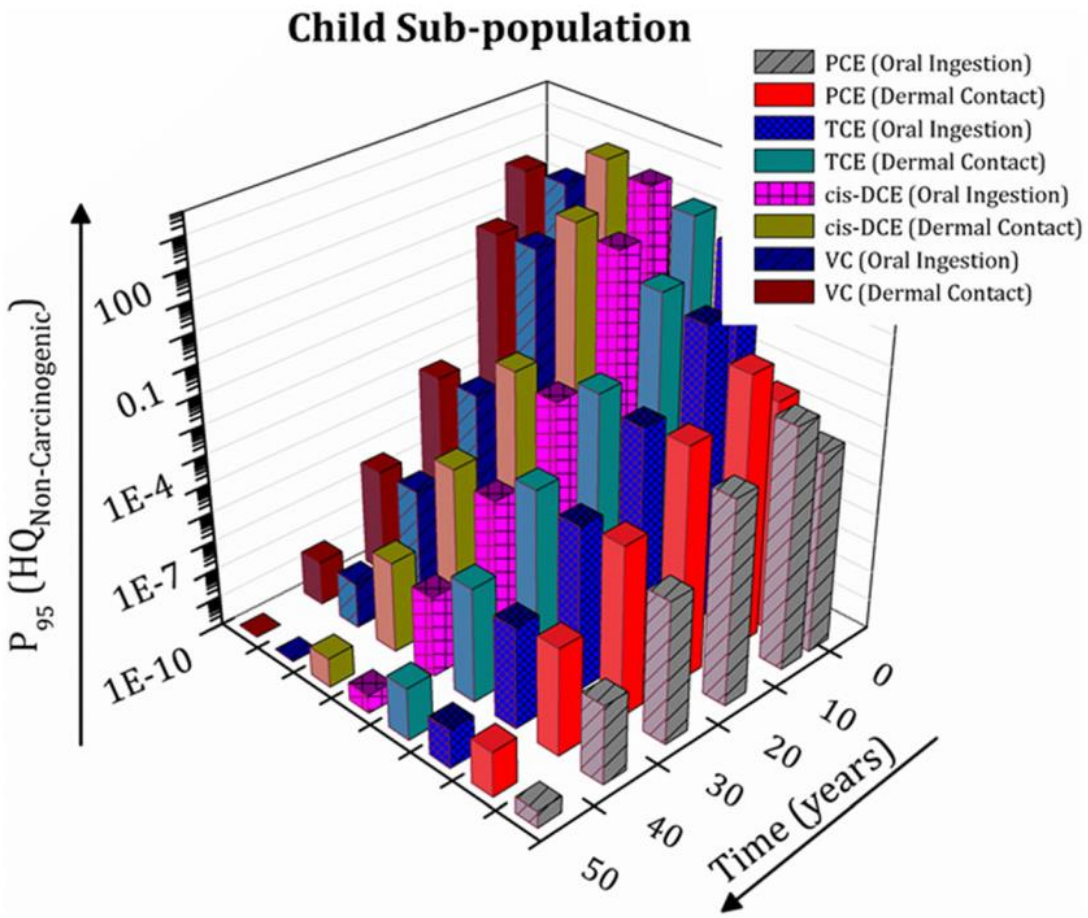


Temporal variation of the mean value of the total (a) non-carcinogenic and (b) carcinogenic health risk

Individual compound effect on non-carcinogenic risk

- cis-DCE pose the highest risk through two exposure routes.





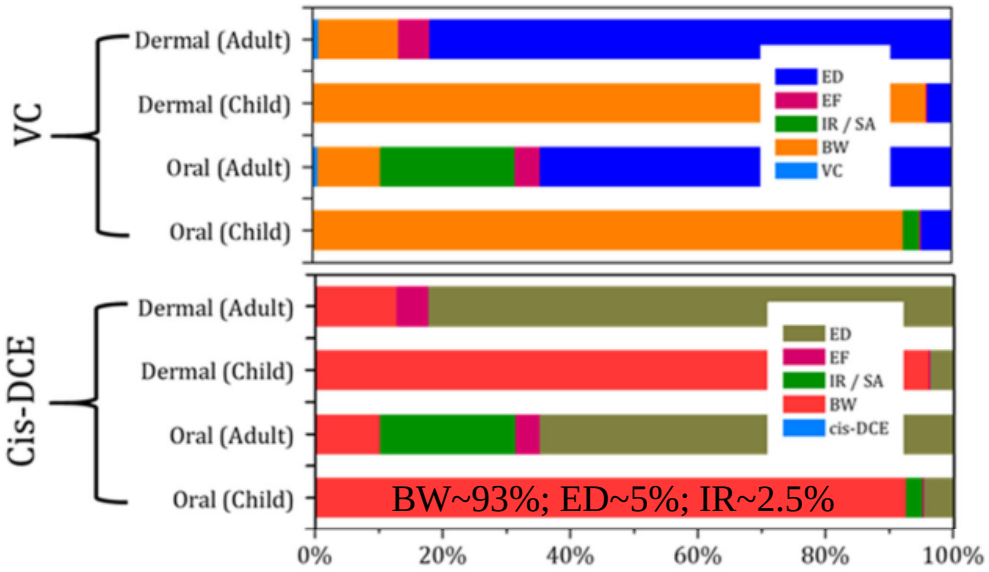
Variance attribution analysis of carcinogenic risk

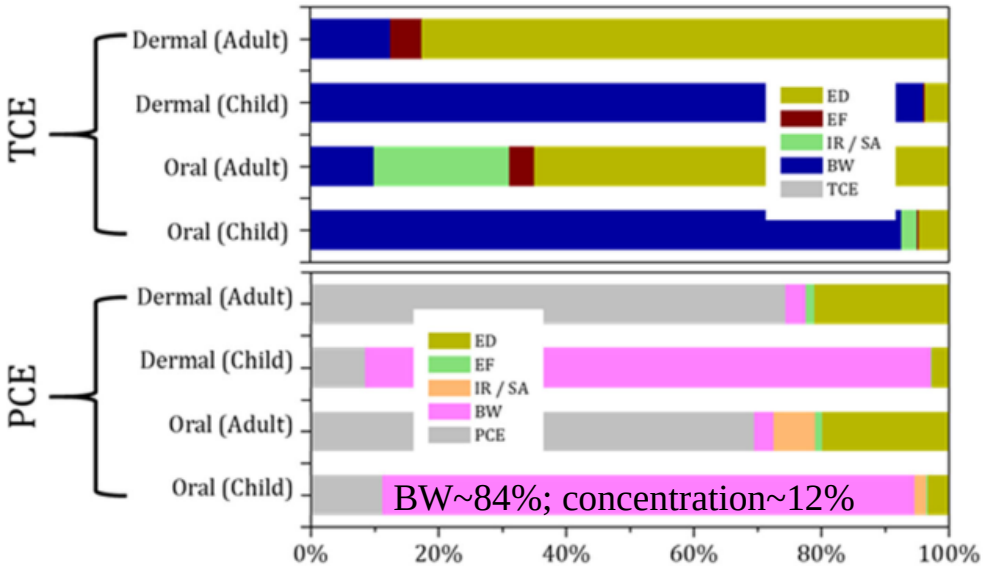
- Contributions of variance of various input parameters are computed as:

$$f_A = \frac{Var(A)\left(\frac{dR}{dA}\right)^2}{\Sigma\left[Var(A)\left(\frac{dR}{dA}\right)^2\right]}$$

Where:
$Var(A)$: variance of the parameter A
f_A: ratio of variance of the parameter A
A include concentration, BW, IR, ED, EF, SA

- Contribution trend varies between PCE and the decay products.
- Contribution also varies between children and adults.





Variance contributions of different parameters for carcinogenic risk estimates at 5th year

Conclusions

- HHRA integrated with the contaminant transport model is an important step in managing a contaminated site and provides a baseline plan to risk managers and authorities for implementing cost and time-efficient remediation woks and guidelines.

- Some findings in this study:
  - VC, cis-DCE pose higher risk in comparison to parent compound (PCE)
  - Bodyweight (BW), concentration, exposure duration (ED), and ingestion rate (IR) were observed as major contributors

- Previous analysis should be included while setting up risk management strategies and in the formulation of remediation measures.

- Risk index computed in this study can be utilized as a useful parameter to make decisions related to remediation management.

Thank you for your attention!

In this work, source dissolution term related to PCE ($s(x,t)$) was neglected. However, source dissolution term was incorporated into the contaminant transport model as an inlet boundary condition in the form of actual on-field dissolved phase concentrations of DNAPL compounds from well situated near to source.

$$(DA_{event})_i = 2 \times FA \times (k_p)_i \times \left(\frac{C_i}{1000}\right) \times \sqrt{\frac{6 \times (\tau_{event})_i \times t_{event}}{\pi}}, \text{for } t_{event} < t^* \quad (4a)$$

$$(DA_{event})_i = FA \times (k_p)_i \times \left(\frac{C_i}{1000}\right) \times \left[\frac{t_{event}}{1+B} + 2 \times \tau_{event}\left(\frac{1+3B+B^2}{(1+B)^2}\right)\right], \text{for } t_{event} > t^* \quad (4b)$$

Where:

FA: fraction absorbed water for contaminant (i)

B: dimensionless ratio of the compound's permeability coefficient through the stratum corneum to its permeability coefficient across the viable epidermis

$(k_p)_i$: dermal permeability coefficient of DNAPL contaminant (i) in water (cm/h)

t_{event}: event duration (hr/event)

$(\tau_{event})_i$: lag time per event for the contaminant (i)

Reference dose and slope factor for DNAPL compound (PCE, TCE, cis-DCE, and VC) via dermal exposure scenario were not available as per our knowledge. Therefore, dermal reference dose value for DNAPL compound was computed by extrapolating the oral reference dose using gastrointestinal absorption factor (ABS_{GI}) (USEPA 2004) as:

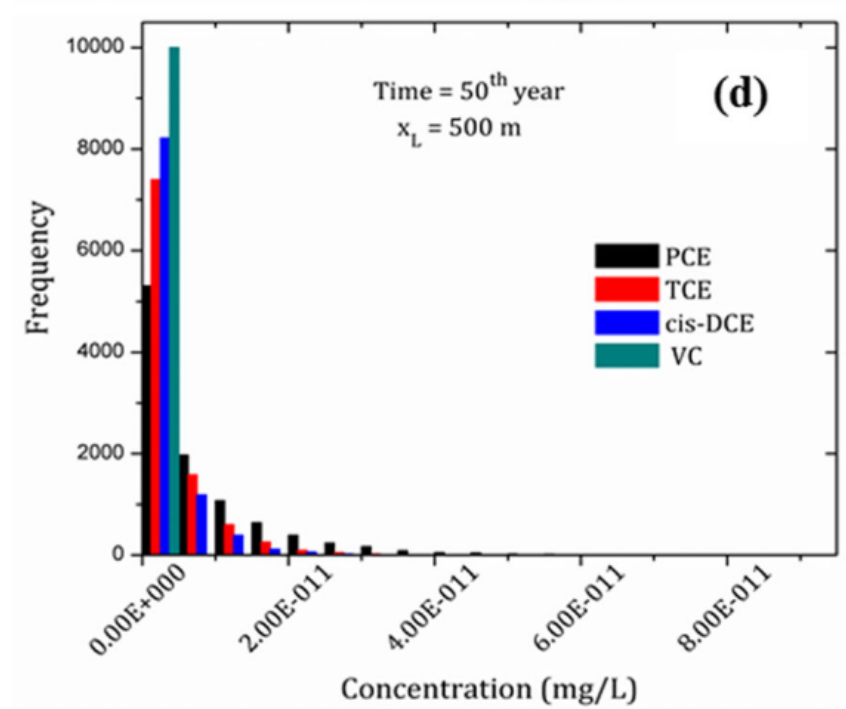
$$RfD_{ABS} = RfD_O \times ABS_{GI} \quad (5)$$

where RfD_{ABS} is the absorbed reference dose (mg/kg-day), RfD_O is the oral reference dose (mg/kg-day), ABS_{GI} is the fraction of contaminant absorbed in the gastrointestinal tract (dimensionless) in the critical toxicity study, and RfD_{ABS}, RfD_O, *and* ABS_{GI} are contaminant specific.

Oral slope factor (RfD_O) were extrapolated using gastrointestinal absorption factor (ABS_{GI}) to get dermal reference dose values (RfD_{ABS}) for DNAPL compound *i* (USEPA 2004) as:

$$SF_{ABS} = \frac{SF_O}{ABS_{GI}} \quad (6)$$

where SF_O is the oral slope factor $(\text{mg/kg-day})^{-1}$ and SF_{ABS} is the dermally adjusted absorbed slope factor $(\text{mg/kg-day})^{-1}$. The value of gastrointestinal absorption factor (ABS_{GI}) = 1 was used for all the DNAPL compounds considered, which was according to USEPA human health evaluation manual (USEPA 2004).



The impact of longitudinal dispersivity on concentration breakthrough curve is analyzed by implementing Monte Carlo type simulations, which could represent the influence of heterogeneity of the porous media on the risk metrics.