ASSESSING THE ROLE OF TRANSPORT MECHANISMS AND MODEL PARAMETERS ON CONTAMINANT PLUME DYNAMICS AND HEALTH RISK METRICS FOR THE SUBSURFACE ENVIRONMENT

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by

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Submitted

in fulfillment of the requirements of the degree of **DOCTOR OF PHILOSOPHY**

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This thesis is dedicated to my beloved parents and my sister

for their endless love, support, and encouragement….

and

to those who are knowingly or unknowingly exposed to

waste-contaminated sites.

CERTIFICATE

This is to certify that the thesis entitled **"Assessing the Role of Transport Mechanisms and Model Parameters on Contaminant Plume Dynamics and Health Risk Metrics for the Subsurface Environment"** submitted by **Abhay** to the Indian Institute of Technology Delhi for the award of the degree of **Doctor of Philosophy** is a bona fide record of research work carried out by him under my supervision. In my opinion, the thesis work has reached the requisite standards, fulfilling the requirements for the degree of Doctor of Philosophy.

The contents of this thesis, in full or in parts, have not been submitted to any other University or Institute for the award of any other degree or diploma.

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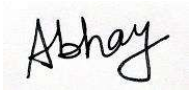

ABSTRACT

Groundwater contamination is increasingly challenging and impacting the ecological environment and human health. To design remediation operations for contaminated regions, contaminant transport models that incorporate uncertainty associated with fate and transport mechanisms need to be developed. Health risk models are necessary to effectively transfer knowledge from technical to non-technical audiences. Moreover, health risk models should include the uncertainty associated with exposure model parameters, site- and activity-specific scenarios and exposure pathways for effective risk communication. This research delves into a multifaceted investigation of contaminant transport in the presence of low permeability porous media (LPPM) and dead-end regions and health risk assessment of the subsurface environment. A probabilistic human health risk assessment (HHRA) framework considering uncertainty in exposure model parameters such as body weight (BW), ingestion rate (IR), exposure frequency (EF), exposure duration (ED), skin surface area (SA) was developed to estimate non-carcinogenic hazard quotient (HQ) due to metal leaching from dumping sites (Okhla, Ghazipur, and Bhalswa landfill sites of Delhi, Ariyamangalam, and fly-ash dumping site Nasik) in India. Fly-ash dumping site in Nasik was found to pose the highest risk to human health in comparison to remaining considered dumping sites. An enormous difference between estimated HQ (child) and HQ (adult) for both the oral ingestion and skin dermal exposure scenarios was not observed. The variance attribution analysis indicated that the BW and ED contributed 35% to 55% towards the overall uncertainties of estimated HQs for children, and more than 95% of the contribution for adults was found to be governed by ED. An integrated ecological and HHRA framework was developed to assess the ecological and human health risks due to chromite ore processing residue (COPR) dumpsites in

Kanpur, India. The multiple exposure scenarios (oral ingestion and skin dermal contact) and multiple pathways (soil and groundwater pathways) along the activity-specific scenarios were considered in the risk assessment framework. The teen playing in mud activity was found in the highest risk zone among all exposure scenarios in all population groups. The average value of HQ was found to be 4.5, which is four orders higher than the safe limit, and the 95th percentile value of cancer risk (CR) was found to be 9×10^{-4} for skin dermal contact exposure scenario via soil pathway. The presence of Cr(VI) and U in groundwater near the Rania site, above the safe limit, was found to pose a significant threat to every population group. The maximum value of CR was found to be 54.9, the highest among all population groups.

A novel Teaching Learning-based Optimization (TLBO) and mobile-immobile (MIM) model-based integrated approach was implemented to simulate the solute transport through heterogeneous short- (30 cm) and long-column (12.5 m) conditions. The MIM model was found to capture the early arrival of solute, peak concentration, and long tailing in the breakthrough curve (BTC) well for conservative solute transport in 12.5 m long highly heterogeneous soil column and reactive solute transport in a 30 cm column-filled with Glendale clay loam soil. Further, the uncertainty associated with flow and transport parameters of multispecies contaminants was quantified using Global Sensitivity Analysis (GSA). GSA of contaminant transport model based on temporal moment-based output indices indicated the complex interplay between different model parameters and their varying influences on different output indices. This research discussed the contrasting plume evolution dynamics observed in porous systems with and without LPPM regions from two-dimensional (2-D) numerical simulations. The contaminant mass recovery and mean residence time were found to be 25% to 50% lower and 25% to 40% higher in the porous system with the LPPM region than in the porous system without it, respectively. In the end,

contaminant transport model-driven probabilistic HHRA was conducted for a chemical mixture of Tetrachloroethene (also known as PCE) and its transformation products. The transformed daughter species i.e., Vinyl Chloride (VC), and cis-Dichloroethene (cis-DCE) were found to pose the highest risk to human health for a longer duration (up to 15 years) than the parent contaminant species i.e., PCE. The BW, concentration, ED, and IR were found to be major contributors to the total variance in the estimated risk metrics. Overall, the probabilistic risk assessment approaches, and numerical methods to analyze contaminant plume dynamics introduced in this thesis can be helpful in understanding the complexities involved in fate and contaminant transport and for policymakers to transfer knowledge from technical to non-technical audiences effectively.

सार

भूजल प्रदूषण तेजी से चुनौतीपूर्ण हो रहा है और पारिस्थितिक पर्यावरण और मानव स्वास्थ्य पर प्रभाव डाल रहा है। दूषित क्षेत्रों के लिए उपचारात्मक कार्यों को डिजाइन करने के लिए, दूषित परिवहन मॉडल विकसित करने की आवश्यकता है जिसमें भाग्य और परिवहन तंत्र से जुड़ी अनिश्चितता शामिल हो। तकनीकी से गैर-तकनीकी दर्शकों तक ज्ञान को प्रभावी ढंग से स्थानांतरित करने के लिए स्वास्थ्य जोखिम मॉडल आवश्यक हैं। इसके अलावा, स्वास्थ्य जोखिम मॉडल में एक्सपोज़र मॉडल मापदंडों, स्थल और गतिविधि विशिष्ट परिदृश्यों और प्रभावी जोखिम संचार के लिए एक्सपोज़र मार्गों से जुड़ी अनिश्चितता शामिल होनी चाहिए। यह शोध कम पारगम्यता वाले झरझरा मीडिया (एलपीपीएम [LPPM]) और मृत-अंत क्षेत्रों की उपस्थिति में दूषित परिवहन की बहुमुखी जांच और उपसतह पर्यावरण के स्वास्थ्य जोखिम मूल्यांकन पर प्रकाश डालता है। शरीर के वजन (बीडब्ल्यू [BW]), अंतर्ग्रहण दर (आईआर [IR]), एक्सपोज़र आवृत्ति (ईएफ [EF]), एक्सपोज़र अवधि (ईडी [ED]), त्वचा की सतह क्षेत्र (एसए [SA]) जैसे एक्सपोज़र मॉडल मापदंडों में अनिश्चितता पर विचार करते हुए एक संभाव्य मानव स्वास्थ्य जोखिम मूल्यांकन (एचएचआरए [HHRA]) ढांचा विकसित किया गया था भारत में डंपिंग साइटों (दिल्ली के ओखला, गाज़ीपुर, और भलस्वा लैंडफिल साइट, अरियामंगलम और फ्लाई-ऐश डंपिंग साइट नासिक) से धातु की लीचिंग के कारण गैर-कैंसरजन्य खतरे के भागफल (एचक्यू [HQ]) का अनुमान लगाने के लिए। शेष मानी जाने वाली डंपिंग साइटों की तुलना में नासिक में फ्लाई-ऐश डंपिंग साइट मानव स्वास्थ्य के लिए सबसे अधिक खतरा पैदा करने वाली पाई गई। मौखिक अंतर्ग्रहण और त्वचा त्वचीय जोखिम परिदृश्यों के लिए अनुमानित एचक्यू (बच्चे) और एचक्यू (वयस्क) के बीच कोई बड़ा अंतर नहीं देखा गया। विचरण एट्रिब्यूशन विश्लेषण से संकेत मिलता है कि बीडब्ल्यू और ईडी ने बच्चों के लिए अनुमानित एचक्यू की समग्र अनिश्चितताओं में 35% से 55% का योगदान दिया, और

वयस्कों के लिए 95% से अधिक योगदान ईडी द्वारा शासित पाया गया। भारत के कानपुर में क्रोमाइट अयस्क प्रसंस्करण अवशेष (सीओपीआर [COPR]) डंपसाइट्स के कारण पारिस्थितिक और मानव स्वास्थ्य जोखिमों का आकलन करने के लिए एक एकीकृत पारिस्थितिक और एचएचआरए ढांचा विकसित किया गया था। जोखिम मूल्यांकन ढांचे में गतिविधि-विशिष्ट परिदृश्यों के साथ कई जोखिम परिदृश्यों (मौखिक अंतर्ग्रहण और त्वचा त्वचीय संपर्क) और कई मार्गों (मिट्टी और भूजल पथ) पर विचार किया गया था। सभी जनसंख्या समूहों में सभी जोखिम परिदृश्यों के बीच मिट्टी की गतिविधियों में खेलने वाले किशोरों को सबसे अधिक जोखिम वाले क्षेत्र में पाया गया। एचक्यू का औसत मान 4.5 पाया गया, जो सुरक्षित सीमा से चार ऑर्डर अधिक है, और मृदा मार्ग के माध्यम से त्वचा त्वचीय संपर्क जोखिम परिदृश्य के लिए कैंसर जोखिम (सीआर [CR]) का 95वां प्रतिशत मूल्य 9×10^{-4} पाया गया। रानिया साइट के पास भूजल में सुरक्षित सीमा से ऊपर Cr(VI) और U की मौजूदगी, हर जनसंख्या समूह के लिए एक महत्वपूर्ण खतरा पैदा करती पाई गई। सीआर का अधिकतम मूल्य 54.9 पाया गया, जो सभी जनसंख्या समूहों में सबसे अधिक है।

विषम लघु- (30 सेमी) और लंबे-स्तंभ (12.5 मीटर) स्थितियों के माध्यम से विलेय परिवहन को अनुकरण करने के लिए एक उपन्यास शिक्षण शिक्षण-आधारित अनुकूलन (टीएलबीओ [TLBO]) और मोबाइल-इमोबाइल (एमआईएम [MIM]) मॉडल-आधारित एकीकृत दृष्टिकोण लागू किया गया था। एमआईएम मॉडल को 12.5 मीटर लंबे अत्यधिक विषम मृदा स्तंभ में रूढ़िवादी विलेय परिवहन और ग्लेनडेल चिकनी दोमट मिट्टी से भरा हुआ 30 सेमी स्तंभ में प्रतिक्रियाशील विलेय परिवहन के लिए ब्रेकथ्रूकर्व (बीटीसी [BTC]) में विलेय पदार्थ के प्रारंभिक आगमन, चरम सांद्रता और लंबी पूंछ को पकड़ने के लिए पाया गया था। इसके अलावा, संतृप्त छिद्रपूर्ण प्रणाली में बहु-प्रजाति के दूषित पदार्थों के प्रवाह और परिवहन मापदंडों से जुड़ी अनिश्चितता को वैश्विक संवेदनशीलता विश्लेषण (जीएसए [GSA]) का उपयोग करके निर्धारित किया गया था। अस्थायी क्षण-आधारित आउटपुट सूचकांकों के आधार पर दूषित परिवहन मॉडल

के जीएसए ने विभिन्न मॉडल मापदंडों और विभिन्न आउटपुट सूचकांकों पर उनके अलग-अलग प्रभावों के बीच जटिल परस्पर क्रिया का संकेत दिया। इस शोध में 2-डी संख्यात्मक सिमुलेशन से एलपीपीएम क्षेत्रों के साथ और बिना छिद्रित प्रणालियों में देखे गए विपरीत प्लम विकास गतिशीलता पर चर्चा की गई। एलपीपीएम क्षेत्र के साथ छिद्रित प्रणाली में संदूषक द्रव्यमान पुनर्प्राप्ति और औसत निवास समय क्रमशः 25% से 50% कम और 25% से 40% अधिक पाया गया, इसके बिना छिद्रित प्रणाली की तुलना में। अंत में, पीसीई [PCE] और उसके परिवर्तन उत्पादों के रासायनिक मिश्रण के लिए दूषित परिवहन मॉडल-संचालित संभाव्य एचएचआरए का संचालन किया गया। परिवर्तित पुत्री प्रजातियाँ (वीसी [VC], सीआईएस-डीसीई [cis-DCE]) मूल (पीसीई [PCE]) संदूषक प्रजातियों की तुलना में लंबी अवधि (15 वर्ष तक) के लिए मानव स्वास्थ्य के लिए सबसे अधिक खतरा पैदा करती पाई गईं। अनुमानित जोखिम मेट्रिक्स में कुल भिन्नता के लिए बीडब्ल्यू, एकाग्रता, ईडी और आईआर को प्रमुख योगदानकर्ता पाया गया। कुल मिलाकर, इस थीसिस में पेश किए गए संभाव्य जोखिम मूल्यांकन दृष्टिकोण और प्रदूषक प्लम गतिशीलता का विश्लेषण करने के तरीके भाग्य और प्रदूषक परिवहन में शामिल जटिलताओं को समझने और नीति निर्माताओं के लिए तकनीकी से गैर-तकनीकी दर्शकों तक ज्ञान को प्रभावी ढंग से स्थानांतरित करने में सहायक हो सकते हैं।

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LIST OF ABBREVIATIONS

1-D	One dimensional
2-D	Two dimensional
2RNE	Two-region non-equilibrium
3-D	Three dimensional
AACi	Average article citation
ADE	Advection dispersion equation
AF	Adherence factor
AT	Averaging time
BA	Bees Algorithm
BTC	Breakthrough curve
BTEX	Benzene, Toluene, Ethylbenzene, Xylene
BW	Body weight
CDI	Chronic daily intake
CF	Contamination factor
CI	Confidence interval
Cis-DCE	cis-Dichloroethene
COM	Center of mass
COPR	Chromite ore processing residue
CPCB	Central pollution control board
MADE	Macro-dispersion experiment
MC	Monte Carlo
MCL	Maximum concentration level
MCS	Monte Carlo simulations
MIM	Mobile-immobile
MNW	Multi-node well
MODFLOW	Modular Three-Dimensional Finite-Difference Groundwater Flow Model
MPNE	Multi processes non equilibrium
MRMT	Multi-rate mass transfer
MRT	Mean residence time
MSW	Municipal solid waste
MT3DMS	Modular Three-dimensional Multispecies Transport Model for Simulation
MTBE	Methyl tert-butyl ether
MTC	Mass transfer correlations
NoA	Number of articles
NSE	Nash-Sutcliffe efficiency
NTPP	Nashik Thermal Power Plant
OpenFOAM	Open-source Field Operation And Manipulation
PCE	Tetrachloroethene

CR	Cancer risk
CTM	Contaminant transport modelling
CTRW	Continuous time random walk
DAD	Dermally absorbed dose
DNAPL	Dense non-aqueous phase liquid
DRA	Dose-response assessment
DYNIA	Dynamic identification analysis
ECC	Estarreja chemical complex
ED	Exposure duration
EE	Elementary effect
EF	Exposure frequency
EV	Event frequency
FAST	Fourier Amplitude Sensitivity Test
FDM	Finite difference method
FEFLOW	Finite Element subsurface FLOW system
FEM	Finite element method
GCW	Groundwater circulation well system
PHT3D	Three-dimensional Reactive Multicomponent Transport Model for Saturated Porous Media
PRA	Probabilistic risk assessment
PRF	Probabilistic risk framework
PRZM	Pesticide root zone model
PSO	Particle Swarm Optimization
PSY	Publication year
RfD	Reference dose
RMSE	Root mean square error
RPCM	Radial point collocation method
RSA	Regional sensitivity analysis
RT3D	Reaction transport code
RWhet	Random–walk based method
RWPT	Random walk particle tracking
SA	Skin surface area
SAFE	Sensitivity Analysis For Everyone
SF	Slope factor
SRCC	Spearman’s rank correlation coefficient

GLUE	Generalized likelihood uncertainty estimation	SUTRA	Saturated-unsaturated transport
GMS	Groundwater modelling system	TBA	Tert-butyl alcohol
GSA	Global sensitivity analysis	TCA	Tetrachloroethane
GWS	Groundwater system	TCE	Trichloroethene
HHRA	Human health risk assessment	TCi	Total number of citations
HI	Hazard index	TDS	Total dissolved solid
HQ	Hazard quotient	TIAA	Tucson International Airport Area
ICET^3	Integrated contaminant elution and tracer test toolkit	TLBO	Teaching-Learning Based Optimization
IFRC	Integrated Field Research Challenge	T-PROGS	Transition Probability Geostatistical Software
IR	Ingestion rate	USDOE	U.S. Department of Energy
IRIS	Integrated Risk Information System	USEPA	United States Environmental Protection Agency
IWAM	Integrated water resource assessment and modelling	VAFB	Vandenberg Air Force Base
KGE	Kling Gupta Efficiency	VC	Vinyl chloride
LPPM	Low permeability porous media	ZTM	Zeroth temporal moment

LIST OF MATHEMATICAL NOTATIONS AND SYMBOLS

$\overline{C^{model}}$	Mean of model simulated concentration	λ_i	First-order degradation coefficient of i^{th} species
$\overline{C^{observed}}$	Mean of observed concentration	μ_i	Mean of the elementary effects
C_{soil_i}	Concentration of target contaminant in soil sample	μ_{lim}	First-order dissolved-phase decay coefficient immobile region
$K_{d_{im}}$	Sorption distribution coefficient in the immobile region	μ_{lm}	First-order dissolved-phase decay coefficient in mobile region
K_{d_m}	Sorption distribution coefficient in the mobile region	μ_n	Normalized temporal moment of n^{th} order
A_i	i^{th} input variable with variance ($Var(A_i)$)	μ_{sim}	First-order sorbed-phase decay coefficient in immobile region
B_n	Metal background/reference concentration	μ_{sm}	First-order sorbed-phase decay coefficient in mobile region
C_0	Injected concentration of solute source	ρ_b	Bulk density of the porous medium
C_i	Concentration of target contaminant in leachate or groundwater	σ_i	Standard deviation of elementary effects
C_{im}	Concentrations in the immobile region	σ_{model}	Standard deviation of model simulated concentration
C_i^{model}	i^{th} model simulated concentration	$\sigma_{observed}$	Standard deviation of observed concentration

$C_i^{observed}$	i^{th} observed concentration	τ_L	Tortuosity factor
C_m	Concentrations in the mobile region	ϵ_p	Liquid volume fraction
C_s	Metal concentration in soil	Δt	Time step
$C_{sk}(x)$	Coefficient of skewness at location x	Δx	Mesh size
$D_{L,i}$	Molecular diffusion coefficient of species diluted in pure liquid	∇p	Pressure difference between inlet and outlet of domain
$D_{e,i}$	Effective diffusion coefficient in saturated porous media	ABS_{GI}	Dimensionless fraction of absorbed contaminant in the gastrointestinal tract
$D_{m,i}$	Molecular diffusion coefficient of i^{th} species in water	ABS	Dimensionless fraction of absorbed contaminant
I_{geo}	Geo-accumulation index	$ADD_i(GW)$	Chronic daily intake for groundwater pathway
J_i	Mass flux vector	$ADD_i(soil)$	Chronic daily intake for soil pathway
$K_{d,i}$	Sorption distribution coefficient of i^{th} species	CV_{model}	Coefficient of variation of model simulated concentration
L_x	Dimension of domain along *x*-direction	$CV_{observed}$	Coefficient of variation of observed concentration
L_y	Dimension of domain along *y*-direction	CF	Contamination factor
$M_0(x)$	Zeroth temporal moment at location x	CSF_{ABS}	Cancer slope factor for skin dermal contact
M_i	Mean of solutions	CSF_O	Cancer slope factor for oral ingestion

M_n	n^{th} order absolute temporal moment	CSF	Cancer slope factor
Q_m	Mass source term	D	Hydrodynamic dispersion coefficient
R^2	Determination coefficient	DA_{event_i}	Absorbed dose per event for groundwater pathway
R_i	Retardation factor of i^{th} species	DA_{event}	Absorbed dose per event
$T_1(x)$	Mean residence time at location x	$DAD_i(GW)$	Dermally absorbed dose for groundwater pathway
$T_2(x)$	Variance of breakthrough at location x	$DAD_i(soil)$	Dermally absorbed dose for soil pathway
T_F	Teaching factor	FA	Fraction of skin in contact with water
T_i	Teacher at i^{th} iteration	IR_{soil}	Soil ingestion rate
$X_{new,i}$	New solution	K	Hydraulic conductivity
$X_{old,i}$	Old solution	L	Total length of numerical domain
$d_i(j)$	Elementary effect determined for i^{th} parameter by using j^{th} sampling point	N	Number of observation points
e_i	Difference between simulated and observed concentrations	OF	Objective function
f_{A_i}	Variance contribution of A_i to the overall variance of the estimated risk index	RfD_{ABS}	Reference dose for absorption
f_A	Variance contribution of A to the overall variance of the estimated hazard quotient	RfD_O	Reference dose for oral ingestion exposure scenario

k_p	Dermal coefficient of permeability of contaminant	RfD	Reference dose
r_i	Random number	SA_{soil}	Skin surface area for soil pathway
t^*	Time to reach a steady-state for DNAPL	$Var(HQ)$	Variance of estimated hazard quotient
t_{event}	Single contact event duration	$Var(RI)$	Variance of estimated risk index
t_{pulse}	Pulse duration	Y	Yield coefficient for transformation from one to another species
v_m	Porewater velocity in the mobile region	f	Fractions of sorption sites that equilibrate rapidly with the mobile region
y_i	Effective yield coefficient for any reactant or product pair representing the mass ratio of species (i) produced to the species ($i-1$) consumed	q	Flowrate
$\alpha_{L,c}$	Dispersivity along longitudinal direction	t	Time
α_L	Longitudinal dispersivity	u	Darcy velocity
$\alpha_{T,c}$	Dispersivity along transverse direction	v	Porewater velocity
α_{TH}	Horizontal transverse dispersivity	$weight_i$	Normalized weight coefficient
α_{long}	Longitudinal dispersivity	x	Spatial coordinate
β_{bias}	Bias ratio	β	Ratio of mobile region to total pore volume

$\gamma_{variability}$	Variability ratio	θ	Total volumetric water content
θ_{im}	Volumetric water content of immobile region	ρ [risk assessment]	Spearman's rho statistic
θ_m	Volumetric water content of mobile region	ω	First-order mass transfer coefficient