Spatio-temporal Indoor Human Exposures in Homes Affected by Chemical-contaminated Soil and Groundwater

Leonid Turczynowicz

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School of Public Health

Faculty of Health and Medical Sciences

University of Adelaide, South Australia

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CONTRIBUTION TO KNOWLEDGE

This research provides new insights concerning indoor air contaminant exposure assessment associated with vapour intrusion (VI).

A summary position from the literature, identifies knowledge gaps. Case studies explore these gaps and provide quantitative data related to volatile organic compound (VOC) concentrations in terms of within-building spatial and temporal variance and the factors that influence that variance. The findings can be used to establish a new evidence-based indoor air sampling strategy that represents 'worst case' conditions, consistent with the public health precautionary principle. Furthermore, this enables an improved understanding of the human health effects of volatile substances from ground contamination and identifies the need to develop a dynamic, time-dependent model of vapour intrusion in buildings. In addition, the research explores a biological monitoring approach, with end-exhaled breath sampling and testing under environmental conditions. This approach shows promise as a non-invasive means of assessing exposure and uptake.

ABSTRACT

INTRODUCTION

Public health problem statement

Vapour intrusion is a process that involves the migration of volatile chemicals from contaminated soil and/or groundwater into dwellings or other confined structures where inhalational exposure may occur. The process is exemplified by naturally occurring soil radon, which is considered a major risk factor for lung cancer in a number of countries. However, the extensive use of industrial chemicals and fuels over many years has left a legacy of soil and groundwater contamination potentially posing an even wider variety of disease endpoints. Volatile substances such as benzene and trichloroethylene (TCE) have also been shown to intrude into buildings from single or multiple sources off site. Limited environmental epidemiological studies have reported increased risks of cancer and non-cancer effects. Owing to the number of people potentially exposed including vulnerable subpopulations, regulatory agencies have developed frameworks for risk assessment based on environmental sampling and predictive models.

These approaches, however, are based on idealized contaminant source and migration

characteristics, and have been found to have limited predictive value for health risk assessment. Further research is required to provide more confidence in risk assessment outcomes.

Initial literature review

In Australia, the federal enHealth's human health risk assessment framework and a National Environment Protection Measure provide the basis for site contamination assessment. These documents, although recognizing the complexity of vapour intrusion, provide limited guidance on exposure assessment and have a focus on petroleum hydrocarbons.

The human health risk assessment of vapour intrusion can be structured into three key areas:

- Sub-surface fate and transport models and vapour measurement that establishes the vapour concentration at the building boundary.
- Ventilation models and measurement which consider indoor air concentrations in space and over time within the building; and
- Human inhalation dosimetry which considers absorbed doses over time.

The peer-reviewed scientific literature on vapour intrusion over the past thirty years combined with the international regulatory documentation is extensive. However, the majority of this literature is oriented towards the initial phase of sub-surface transport to the building boundary. There has been limited focus on ventilation dynamics and less so on inhalation dosimetry. In the past five years, however, increasing attention has focused on spatial and temporal indoor air contaminant changes with one public health publication on residential indoor air spatio-temporal variability and another on linking indoor air contaminant concentrations to biological markers. In terms of considering inhalation dosimetry, however, there is as yet, no discourse on how these indoor environments may result in differing inhalation doses. This may be particularly important where high level peak doses due to environmental effects on the distribution of an indoor volatile, result in adverse pathologies. Indeed, there is some evidence, for example, in the case of TCE, for peak tissue concentrations precipitating acute neurotoxic effects.

Gap in knowledge

The gaps in knowledge in vapour intrusion exposure assessment include the following:

- Models of dynamic and time-dependent (non-steady state) vapour migration processes and their validation.
- An understanding of spatio-temporal variability in indoor concentrations and the correlates of the variability that might lead to an evidenced-based indoor air sampling protocol.
- The time dependence of absorbed dose, especially tissue concentrations, that results from time-dependent inhaled air concentrations.

PURPOSE STATEMENT

Through a critical review of the literature and a series of empirical case studies, this research seeks to:

- Elucidate the nature of spatial-temporal changes in indoor contaminant concentrations within houses affected by vapour intrusion and the factors that may influence those changes.
- Provide an evidence base for a time-dependent vapour intrusion model with empirical evaluation, applicable to Australian conditions.
- Explore the utility of biological monitoring for risk assessment in a common vapour intrusion scenario.

GENERAL RESEARCH QUESTIONS

- What is the short- and long-term spatio-temporal variability of indoor air contaminants arising from vapour intrusion?
- Which factors are significantly associated with indoor air concentration variability?
- What is the relationship between biological monitoring data and indoor TCE concentrations?

METHODS

A critical literature review and experimental case study approach were used. The experimental case studies were opportunistic and reflected real-life conditions. Case Study 1 was a termiticide treatment (including xylene) in a suspended floor home and Case Study 2 was a slab-on-ground house in a TCE-affected area.

Critical literature review

Computerised searches of the published literature were conducted using the Web of Science, Scopus and PubMed. The logic grid included "*vapour intrusion*"; "*ventilation*"; "*inhalation dosimetry*" and "*exposure*". The yields were complemented with author searching and forwards and backwards searching. The literature on vapour intrusion was critically reviewed in terms of its utility for human health risk assessment.

Case Study 1 – Suspended timber floor home construction - Indoor air concentrations

The upper portion of the soil in the subfloor of a 1950's home was treated with technical grade xylene containing m-, p- and o-xylenes as part of a termiticide treatment. Analyses were conducted of soil xylene and moisture concentrations; subfloor and indoor air xylene concentrations; and air exchange rates in the subfloor space and occupied space. Concurrent meteorological data were collected from a weather station. A published Australian non-steady state model, developed in previous national guidance, was used to estimate (and compare with) indoor air concentrations based on the empirical measurements.

Case Study 2a – Concrete slab on ground home construction - Indoor air concentrations

A four-bedroom public housing property in a residential area impacted by chlorinated hydrocarbon contaminated groundwater was used over a period of 14 months to assess indoor air TCE levels. Passive TCE sampling occurred at five indoor and two outdoor locations over various time intervals. Air exchange rates were calculated at front and rear indoor sampling locations. Detailed local meteorological data were gathered from a weather station. Indoor temperature and indoor relative humidity were measured at 30 minute intervals over a 3-month period at each of five indoor air sampling locations. Soil vapour, sub-slab vapour and flux chamber measurements were carried out during

one week concurrent with 6 hour passive sampling.

Case Study 2b - Slab on ground home construction - Human exposure experiments:
A biological monitoring pilot study was conducted with 5 volunteer adults who occupied the TCE-contaminated house for 12 hours. End-exhaled breath samples and blood samples were collected. Participants were also asked to provide urine samples at baseline, at the end of the exposure period and on three subsequent occasions. Passive indoor air sampling and surface flux testing was undertaken. Sub-slab TCE samples were also collected inside and outside the house.

RESULTS

Critical literature review

Papers on vapour intrusion mainly focused on issues associated with the sub surface. These included areas such as development of one- and three-dimensional steady-state models; estimation of attenuation factors; lateral exclusion distances; factors affecting subsurface migration such as moisture levels and oxygen concentrations and reconsideration of the United States (US) Environment Protection Agency vapour intrusion database. There have, however, been some new areas of focus in the last five years, which have included the use of new real-time measurement techniques; an increased focus of the role of pressure differences on indoor air contaminant concentrations; seasonal and diurnal differences and spatio-temporal variability in homes across an affected community. One recent study examined indoor air TCE concentrations and blood TCE levels. The recent literature has increasingly examined the above-ground and indoor environment but has not further considered within-building spatial differences nor a more detailed examination of short-term indoor air average concentration changes and associated influencing variables. In addition, the literature is silent on the issue of inhalation dosimetry in vapour intrusion and the potential for non-invasive methods of biological monitoring.

Case Study 1– Suspended floor home construction - Indoor air concentrations

Xylene air concentrations decayed to non-detectable levels within two weeks. Subfloor xylene air concentrations were greater than living space xylene air concentrations, and the decay of the concentrations following a generally consistent pattern. Air exchange rates between the sub-floor and living space differed by up to an order of magnitude and demonstrated the influence of subfloor ventilation on vapour intrusion. Statistically significant associations were found for air exchange in the sub-floor space and locally measured minimum and average wind speed.

Site-specific variables in a non-steady state model showed general consistency with measured data, but the modelling estimated a greater shorter-term initial peak with more rapid decay of xylene concentrations than those measured.

Case Study 2a - Slab on ground home construction - Indoor air concentrations
Air sampling data revealed spatial and seasonal indoor TCE variations. Winter month results were up to an order of magnitude greater than summer months. Monitoring over 6-hour (h) periods demonstrated the occurrence of diurnal peaks that were not evident with a 24-h sampling regime. Moreover, the use of a continuous data logging instrument showed occasional spikes over rapid time intervals which were an order of magnitude or greater compared to the common baseline value. Air exchange measurements revealed consistent early morning declines in ventilation. Correspondingly, the highest surface TCE flux was noted during the day with the lowest occurring during the evening. Soil vapour measurements at progressive depths at the rear of the property showed high source concentrations of TCE with lower concentrations progressively up to the sub-slab.

Using the 6-h average TCE concentration as the outcome variable, it was found that ventilation, internal temperature, barometric pressure and wind direction were significant predictor variables in a multivariate model. Ventilation had the greatest impact in the best fit model with one air change per hour predicting a 4.4 $\mu g\ m^{-3}$ decline in the indoor TCE concentration. Assessment of model predictions showed close agreement with the dataset.

Case Study 2b - Slab on ground home construction - Human exposure experiments
The pilot biological monitoring exercise yielded mixed results with most biomonitoring data below the limit of reporting (LOR) which was which was $<5\mu g\ m^{-3}$ for breath and $<0.01\ \mu g\ L^{-1}$ for blood. End-exhaled breath TCE concentrations were generally below $5\mu g\ m^{-3}$ with two results above the LOR. Composite end-exhaled breath samples for baseline and at 02:30 and 08:30 were 2.0, 1.5 and $1.2\mu g\ m^{-3}$ respectively. Blood concentrations were all below the level of reporting of $0.01\ \mu g\ L^{-1}$. While blood TCE concentrations could not be quantified in accordance with standard protocols, discrete peaks were observed on the chromatograms.

CONCLUSIONS AND RECOMMENDATIONS

The recent literature has increasingly examined the above-ground and indoor environment but has not further considered within-building spatial differences nor short-term indoor air average concentration changes and their influencing variables. In addition, the literature is silent on the issue of inhalation dosimetry in vapour intrusion and the potential for non-invasive methods of biological monitoring.

Case Study 1 confirmed the influence of dwelling features and that of ventilation and meteorological variables such as wind speed for a suspended-floor dwelling.
Case Study 2 captured greater resolution across all measurements and although the extent of the variables measured varied, sufficient data were captured to provide a more detailed examination of time-dependent change. Statistically significant spatial differences were observed suggesting the need to account for prevailing wind direction in worst case indoor sampling strategies. Mixed-effects regression models were consistent with the observed seasonal and diurnal differences. The two case studies provide evidence for a worst-case sampling strategy, that is, sampling in winter and during the evening and accounting for spatial variance.

Overall, the results demonstrate the complexity of indoor ventilation dynamics and that spatial and temporal influences are important to understand for exposure assessment purposes. Short term, peak TCE exposure periods were observed and may be of toxicological significance based on information suggesting TCE exhibits a non-monotonic dose-response relationship for foetal malformations.

On the basis of the research the following recommendations are made:

- More detailed and extensive (>1 year) longitudinal studies capturing time-dependent changes in indoor air concentrations and all influencing variables including air pressure changes, should be undertaken.
- A human volunteer biological monitoring study using end-exhaled breath and blood TCE analyses should be undertaken, using sensitive analytical techniques such as Selected Ion Flow Tube Mass Spectrometry.
- A retrospective epidemiological study in TCE-affected areas should be conducted in Adelaide.

DECLARATION

I certify that this work contains no material which has been accepted for the award of any other degree or diploma in my name, in any university or other tertiary institution and, to the best of my knowledge and belief, contains no material previously published or written by another person, except where due reference has been made in the text. In addition, I certify that no part of this work will, in the future, be used in a submission in my name, for any other degree or diploma in any university or other tertiary institution without the prior approval of the University of Adelaide and where applicable, any partner institution responsible for the joint-award of this degree.

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I acknowledge the support I have received for my research through the provision of an Australian Government Research Training Program Scholarship.

Signed: _____

Date: _____

Leonid Turczynowicz BAppSc MPH
Adelaide, South Australia

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LIST OF ABBREVIATIONS, ACRONYMS AND SYMBOLS

θ_G	Air-filled porosity or volumetric air content
α	Alpha = attenuation factor (AF)
~	Approximately
μT	Degradation rate constant of contaminant in soil
μ_a	Degradation rate constant of contaminant in air
^oC	Degrees Centigrade
ρ_B	Dry bulk density
4-h	Four hour
>	Greater than
1_IN	Location one inside also referred to as R1
2_IN	Location two inside also referred to as R2
3_IN	Location three inside also referred to as R3
4_IN	Location four inside also referred to as R4
5_IN	Location five inside also referred to as R5
6_OUT	Location six outside
$\mu g\ L^{-1}$	Micrograms per litre
$\mu g\ m^{-3}$	Micrograms per cubic metre
$\mu g\ m^{-2}\ min^{-1}$	Micrograms per square metre per minute
ε	n-by-1 observation error vector
1-D	One-dimensional
β	p-by-1 fixed-effects vector
ρ_P	Particle density
%	Percentage
% v/v	Percentage by volume
®	Registered trademark
7-d	Seven day
7_OUT	Radiello location 7 outside
6-h	Six hour
6_OUT	Radiello location 6 outside
3-D	Three-dimensional
ϕ	Total soil porosity
3-D	Three-dimensional
12-h	Twelve hour
24-h	Twenty-four hour
Ω_D	Volume of interior of the house
Ω_{CS}	Volume of subfloor space
θ_L	Water-filled porosity or volumetric water content
A	Area
ACH	Air changes per hour
ACH_1_IN	Ventilation variable for air exchange at location R1/1_IN (6-h average)
ACH_1_IN30min	Ventilation variable for air exchange at location R1/1_IN (30min average)
ACH_4_IN	Ventilation variable for air exchange at location R4/4_IN (6-h average)

LIST OF ABBREVIATIONS, ACRONYMS AND SYMBOLS (CONTD.)	
ACH_4_IN30min	Ventilation variable for air exchange at location R4/4_IN (30min average)
AEA	Australian Environmental Auditors Pty Ltd
AF	Attenuation factor
AHTA	Adelaide Health Technology Assessment
AMTB	Air-Mucous-Tissue-Blood
ASC-NEPM	Australian Site Contamination National Environment Protection Measure
ASL	Arterial Spin Labelling
AS/NZS	Australian Standard/New Zealand Standard
ASTM	American Society for Testing and Materials
ATSDR	Agency for Toxic Substances and Disease Registry
AUC	Area Under Curve
AVE RH	Average relative humidity
AVE temp ^{o}C	Average temperature degrees celsius
AVE WD	Average wind direction
b	q-by-1 random-effects vector
BOM	Bureau of Meteorology
bp	Barometric pressure variable
BRAC	Base Realignment and Closure (Act)
BSI	British Standards Institute
BTEX	Benzene, Toluene, Ethyl benzene and Xylene
CARACAS	Concerted Action on risk Assessment for Contaminated Sites in the European Union
bp	Barometric pressure variable
c	Concentration
CCME	Canadian Council of Ministers of the Environment
CFD	Computational Fluid Dynamic
CFD-PBPK	Computational Fluid Dynamic-Physiologically-Based Pharmaco-Kinetic
CI	Confidence Interval
CIHD	Cumulative Indoor Human Dose
CIRIA	Construction Industry Research and Information Association
CL:AIRE	Contaminated Land: Applications in Real Environments
CLEA	Contaminated Land Exposure Assessment model
CLM Act	Contaminated Land Management Act
cm	Centimetre
cm^3	Centimetre cubed
$cm^3\ cm^{-3}$	Centimetre cubed per centimeter cubed
C_{max}	Maximum concentration
C_0	Initial concentration of soil contaminant
CO_2	Carbon Dioxide
CO	Carbon Monoxide
COPC	Chemicals of Potential Concern
CPM	Controlled Pressure Method
CRC CARE	Cooperative Research Centre for Contamination and Remediation of the Environment

LIST OF ABBREVIATIONS, ACRONYMS AND SYMBOLS (CONTD.)	
CSIRO	Commonwealth Scientific and Industrial Research Organisation
CSM	Conceptual Site Model
CVI	Chemical Vapour Intrusion
C x t	Concentration multiplied by time
d_{air}	Depth of the soil/atmosphere boundary layer
D_G^{air}	Volatile diffusivity in air
D_L^{water}	Volatile diffusivity in water
Day^{-1}	Per day
DCE	Cis-1,2-dichloroethylene/trans-1,2-dichloroethylene
DEFRA	Department for Environment, Food and Rural Affairs
deg	Degrees
DER	Department of Environmental Regulation
DHHS	Department of Health and Human Services
DoD	Department of Defence
DT	Effective diffusion co-efficient
DTSC	Department of Toxic Substances Control
DV	Dependent Variable
E	East
EC	Equivalent Carbon
ECOS	Environmental Council of the States
EDC	Endocrine Disruptor
EEA	European Environment Agency
EHRA	Environmental health risk assessment
EPA	Environment Protection Authority or Agency
Epi	Epithelium
ERIS	Environmental Research Institute of the States
ESCALE	Etude Sur les Cancers et les Leucémies de l'Enfant
EU	European Union
F2	A notation for twice the stimulation frequency
F5	Flux sampling location 5
f_{oc}	Organic carbon fraction
F_{out}	Flux monitoring location outside
FRC	Functional Residual Capacity
FRLI	Federal Register of Legislative Instruments
FUDS	Formerly Used Defence Sites
g/100g	Grams per hundred grams
$g\ m^{-3}$	Grams per cubic metre
GC/ECD	Gas chromatography/Electron Capture Detection
GC/FID	Gas Chromatography/Flame Ionisation Detection
GC/MS	Gas chromatography/Mass Spectrometry
HI	Hazard Index
g/100g	Grams per hundred grams
HIL	Health Investigation Levels
HNF4a	A transcription factor

LIST OF ABBREVIATIONS, ACRONYMS AND SYMBOLS (CONTD.)	
HQ	Hazard Quotient
I	Average air exchange rate
IABR	International Association for Breath Research
IARC	International Agency for Research on Cancer
IAQ	Indoor Air Quality
Indoor_Temp_0C	Indoor temperature variable
IPCS	International Programme on Chemical Safety
IRIS	Integrated Risk Information System
ITRC	Interstate Technology and Regulatory Council
J&E	Johnson and Ettinger
JEM	Johnson and Ettinger Model
JRC	Joint Research Centre
K_D	Distribution coefficient
kg	Kilogram
K_H	Henry's Constant
K_{OC}	Organic carbon partition coefficient
L	Depth (of contamination in modelling)
L	Litre
LBW	Low Birthweight
LEL	Lower Explosive Limit
LME	Linear Mixed Effects
lnC	Natural logarithm of carbon dioxide concentration
Location	Location variable (R1/1_IN to R5/5_IN)
LOR	Limit of Reporting
m	Metre
m^2	Metre squared
m^3	Metre cubed
$m\ d^{-1}$	Metres per day
mBar	Millibar
AHTA	Massachusetts Department of Environmental Protection
MfE	Ministry for the Environment
$m\ g^{-1}$	Metre cubed per gram
$m^2\ d^{-1}$	Metre squared per day
$m^3\ g^{-1}$	Metre cubed per gram
$m^3\ m^{-3}$	Metre cubed per metre cubed
$mg\ m^{-3}$	Milligram per cubic metre
$mg\ kg^{-1}\ d^{-1}$	Milligram per kilogram per day
ml	Millilitre
mm	Millimetre
ms	Millisecond
$m\ s^{-1}$	Metres per second
MTBE	Methyl-t-butyl ether
N	North
NASA	National Aeronautical Space Administration

LIST OF ABBREVIATIONS, ACRONYMS AND SYMBOLS (CONTD.)	
NATA	National Association of Testing Authorities
NE	North-east
NEE	North-east-east
NES	National Exposure Standard
NEPC	National Environment Protection Council
NEPM	National Environment Protection Measure
ng	nanogram
NIOSH	National Institute of Occupational Safety and Health
NJDEP	New Jersey Department of Environment Protection
NMDR	Non-Monotonic Dose Response
NNE	North-north-east
NNW	North-north-west
NRC	National Research Council
NSW	New South Wales
NSW EPA	New South Wales Environment Protection Authority
NSW DECCW	New South Wales Department of Environment, Climate Change and Water
NT EPA	Northern Territory Environment Protection Authority
NW	North-west
NWW	North-west-west
NYSDoH	New York State Department of Health
O_2	Oxygen
OECD	Organisation for Economic Co-operation and Development
OEH	Office of Environment and Heritage
OEH laboratory	Occupational and Environmental Hygiene laboratory
OR	Odds Ratio
OSWER	Office of Solid Waste and Emergency Response
Outdoor_Temp_0C	Outdoor temperature variable
PBPK	Physiologically-Based Pharmaco-Kinetic
PCE	Perchloroethylene also known as tetrachloroethylene
PID	Photo-ionisation Detector
Po-214	Polonium-214 isotope
Po-218	Polonium 218 isotope
ppb	Parts per billion
ppm	Parts per million
PTFE	Polytetrafluoroethylene
Q_{CD}	Volumetric flow rate from subfloor to dwelling interior
QLD EPA	Queensland Environment Protection Authority
R1	Radiello location 1 inside, same as 1_IN
R2	Radiello location 2 inside, same as 2_IN
R3	Radiello location 3 inside, same as 3_IN
R4	Radiello location 4 inside, same as 4_IN
R5	Radiello location 5 inside, same as 5_IN
Relative_Humidity	Relative humidity variable
Resultc	Passive sampling results corrected for outdoor concentrations
RfC	Reference Concentration

LIST OF ABBREVIATIONS, ACRONYMS AND SYMBOLS (CONTD.)	
RfD	Reference Dose
RAGS	Risk Assessment Guidance for Superfund
RCRA	Resource Conservation and Recovery Act
R_G	Partition coefficient
RI	Response Index
RIVM	Dutch National Institute for Public Health and the Environment
RR	Rate ratio
RT	Response Time
RT_{100}	100ms increase in Response Time
s	Second
S	South
SA EPA	South Australian Environmental Protection Authority
Sagittal	Sagittal plane which divides the body into left and right
SAQP	Sampling and Analysis Quality Plan
SE	South-east
SGV	Soil guideline values
SI	Sensitivity Index
$SI_{0.1}$	0.1 unit decrease in Sensitivity Index
Solar_radiation	Solar radiation variable
SS3	Sub-slab location 3
SS4	Sub-slab location 4
SSE	South-south-east
SSW	South-south-west
STEL	Short-Term Exposure Limit
Sub	Submucosal tissue
SVQGIAQ	Soil Vapour Quality Guidelines for Indoor Air Quality
SWW	South-west-west
SV3-2m	Soil vapour location 3 at 2m depth
SV3-7m	Soil vapour location 3 at 7m depth
SV3-2m	Soil vapour location 3 at 14m depth
t	Time
t_1	Time one
t_2	Time two
TAS EPA	Tasmanian Environment Protection Authority
TCA	Trichloroethane
TCA	Tolerable Concentration in Air
TCAA	Trichloroacetic acid
TCE	Trichloroethylene
TCOH	Trichloroethanol
™	Trademark
TPH	Total Petroleum Hydrocarbons
TPHCWG	Total Petroleum Hydrocarbon Criteria Working Group
TRV	Toxicological Reference Value
TWA	Time-Weighted Average

LIST OF ABBREVIATIONS, ACRONYMS AND SYMBOLS (CONTD.)	
UK	United Kingdom
UPSS	Underground Petroleum Storage Systems
US	United States
USA	United States of America
US DOD	United States Department of Defence
US EPA	United States Environmental Protection Agency
UST	Underground Storage Tank
V_L^B	Volumetric soil water flux
VIF	Variance Inflation Factor
VC	Vinyl Chloride
VEP	Visual Evoked Potential
VI	Vapour Intrusion
Vic EPA	Victorian Environment Protection Authority
VEP	Visual Evoked Potentials
VOC	Volatile Organic Compound
V_T	Effective solute velocity
W	West
WA EPA	West Australian Environment Protection Authority
WCA	WorkCover Australia
WHO	World Health Organisation
Wind_direction	Wind direction variable
Wind_directionC	Wind direction variable, by quadrant of 22.5º each (16 quadrants)
W m^{-2}	Watts per square metre
X	n-by-p fixed-effects design matrix
X_{CS}	Ventilation rate of subfloor space
X_D	Ventilation rate of dwelling interior
y	n-by-1 response vector
Z	n-by-q random-effects design matrix

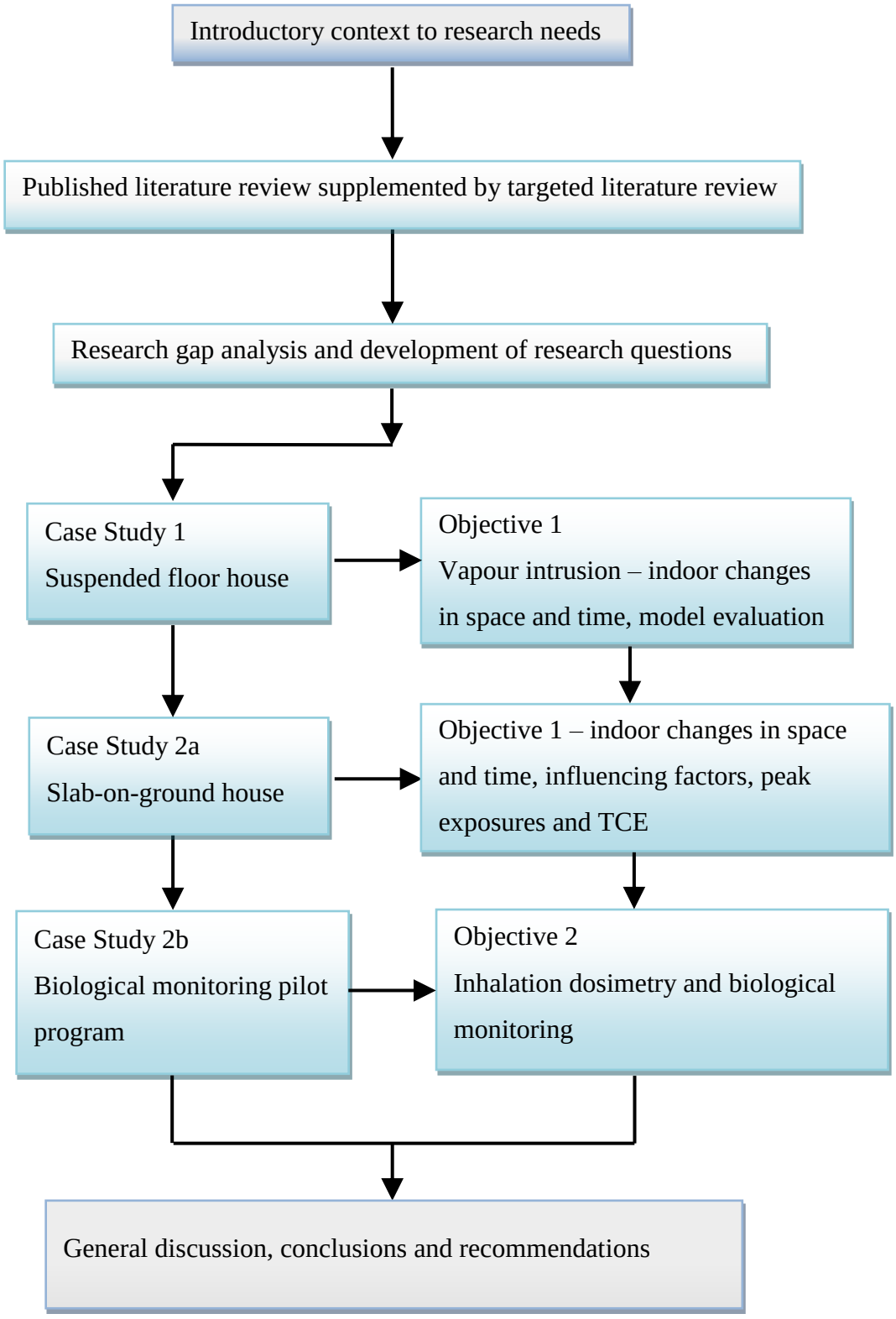
LIST OF APPENDICES

THESIS OVERVIEW

The purpose of this overview is to provide a summary of the key elements of the individual chapters of the thesis. The thesis has been structured in a traditional sense with an introduction followed by a literature review, research questions, methodology, empirical research findings, discussion, conclusions and recommendations. A case study approach has been used, providing an increased understanding and direction for the experimentation undertaken, based on the premise that time-dependence is a key factor in understanding exposure assessment. This commenced with a preliminary program investigating xylene application and site-specific evaluation of an Australian non-steady-state vapour intrusion model in a suspended floor dwelling. This was followed by a more detailed experiment from the knowledge gained which examined a real-life setting where soil and groundwater TCE contamination had impacted a house resulting in occupant evacuation. The empirical investigation included both environmental and biological monitoring and produced information which can lead to an indoor sampling protocol and a non-invasive biological monitoring method.

This thesis comprises six chapters, with a flow diagram of the structure presented in the following figure and the content and key messages presented in the subsequent table.

THESIS OVERVIEW FIGURE



THESIS OVERVIEW TABLE

Chapter	Chapter content	Key messages
1-Introduction.	This chapter provides the background to the international problem of vapour intrusion. It highlights the human health risk assessment process and the public health significance.	Vapour intrusion is a global public health problem associated with site contamination. Its assessment involves the determination of indoor air concentrations in houses above or near contamination. This is achieved through modelling or measurement. Both methods have significant knowledge gaps requiring further development.
2-Literature review.	The literature search objectives are described and search strategy presented. The search outcomes are summarised to present the gaps in knowledge pertaining to spatial and temporal understanding of indoor air exposure from vapour intrusion and current regulatory frameworks. A rationale for the proposed research is presented and the aims and research questions detailed arising from the literature review. Out of scope research aspects are presented.	Vapour intrusion literature is oriented towards subsurface concerns with limited focus on the indoor environment and the inhalation dose. Spatial and temporal concerns more recently raised, highlight the uncertainties, but within-building spatial distribution and temporal aspects of inhalation and dose are not considered. Public health focus is limited with only two papers providing a community impact perspective. Closer examination of these aspects is warranted.
3-Case Study 1. Evaluation of a suspended floor house.	Xylene application to the surface of a suspended floor home was monitored for above-ground xylene decay with ventilation and meteorological factor assessment. An Australian non-steady-state model was evaluated by capturing the necessary site-specific variables.	Xylene diminished in subfloor and indoor air within two weeks. Subfloor xylene levels were greater than indoor air levels with a consistent decline in decay reflected by an order of magnitude difference in air exchange between compartments. Wind speed was associated with subfloor air exchange rates. Diurnal effects were noted with the use of a PID. The model evaluation reported a greater and faster peak and more rapid decline than empirical data suggesting an influence of the diurnal effect. Higher resolution experimentation was a subsequent requirement.
4-Case Study 2. Evaluation of a slab-on-ground house.	A slab-on-ground vacated 4-bedroom home in a TCE affected area was monitored over a period of 14 months using passive sampling methods of varying time-averages (4h, 6h, 12h, 24h and 7d) and durations with concurrent capture of a range of environmental variables including ventilation.	Spatial and temporal changes were observed and seasonal and diurnal effects reported. Indoor air TCE levels were statistically associated with a range of environmental factors. Multivariate modelling reported the most significant factors in order of significance as ventilation, wind direction, barometric pressure and internal temperature.

Chapter	Chapter content	Key messages
4-Case Study 2. Evaluation of a slab-on-ground house (contd).	A short-term human exposure study of 12 hours with biological monitoring explored TCE in end-exhaled breath, blood and urine in five participants.	Biological monitoring showed mixed results due to a decline in the indoor air TCE concentrations but composite end-exhaled breath and indoor air averages showed general consistency with published *f* values. Blood TCE concentrations were below the LOR of 0.01µg/L but TCE peaks were reported on chromatograms.
5-General discussion.	The novelty and significance of the research is highlighted and findings discussed in the context of international literature. Generalisability is discussed and the strengths and limitations explored.	The public health focus has been on exposure assessment hence the assessment of within-building spatial and temporal variability was the key determinant not considered elsewhere. The qualitative and quantitative nature of this evaluation enabled statistical model development. While the findings are supported on the qualitative level the literature is limited on the quantitative level. For example, the quantitative relationships of the significant predictors for indoor TCE concentrations and the evidence of intermittent short-term peak elevations in TCE exposure. The latter potentially being of consequence for a substance exhibiting a non-monotonic dose-response relationship. End-exhaled breath TCE sampling and analysis show promise for environmental biological monitoring.
6-Conclusions and Recommendations	Conclusions from the research findings are presented with recommendations for researchers, government and industry.	The application of this knowledge will improve confidence in exposure assessment. This enables development of an evidence-based "worst case" indoor air sampling framework which is currently unavailable. Recommendations include extension research work with further time-dependent data capture over a year or longer; a biological monitoring study using end-exhaled breath with more robust methods and a retrospective epidemiological study in TCE-affected areas of Adelaide.

General Research Questions

The following questions were developed from the literature review and include:

What spatial and temporal changes are associated with indoor inhalation exposures arising from vapour intrusion and what factors may influence that change?

What inhalation dosimetry understanding is required and what methods of exposure assessment for vapour intrusion need to be considered?

These questions comprise a series of sub-questions structured across three phases of investigation:

- Critical literature review
- Case Study 1 – suspended floor dwelling
- Case Study 2 – slab-on-ground floor dwelling

Each of these sub-questions are raised at the start of each chapter and addressed at the end of each chapter. They are detailed below.

Specific research sub-questions

Critical literature review

RQ1 How important is it to know the variance in spatial and temporal indoor air concentrations for exposure assessment?

RQ2 How relevant is time-dependent vapour intrusion modelling and exposure assessment to health risk assessment?

RQ3 What are the key elements in the current Australian and international regulatory status on this issue?

In the case of a suspended floor dwelling design:

RQ4 How do subfloor and indoor air exchange rates impact on sub-floor and indoor air concentrations?

RQ5 What are the implications associated with sub-floor housing design for vapour intrusion?

RQ6 Is there a relationship between wind speed and air exchange rate?

RQ7 Is there a relationship between wind speed and temperature?

RQ8 Is there a relationship between wind speed and relative humidity?

RQ9 Is there a difference between the local weather station readings and the Bureau of Meteorology (BOM) station?

RQ10 How does temporality compare with the literature?

RQ11 What assessment protocols (time-averaged or continuous) proved most useful?

RQ12 Is the model output consistent with empirical observations?

RQ13 What were the trends in the empirical measurements of indoor air concentrations?

RQ14 How do the measurements support non-steady-state vapour model improvements?

RQ15 What were the key findings in Case Study1?

In the case of a slab-on-ground dwelling design:

RQ16 Are there any significant differences across indoor testing locations?

RQ17 Are there any differences across seasons?

RQ18 Are there any diurnal differences?

RQ19 Are short-term peak exposures observed?

RQ20 Are short-term peak exposures masked by longer-term average measurements?

RQ21 Which environmental factors were significantly associated with the indoor concentrations?

RQ22 Which environmental factors had the greatest influence on indoor TCE concentrations?

In terms of the biological monitoring pilot program:

RQ23 What are reported TCE concentrations in end-exhaled breath?

RQ24 What are TCE concentrations in blood samples?

RQ25 What indoor TCE concentrations are observed?

RQ26 Are TCE concentrations in end-exhaled breath and indoor air consistent with published data?

RQ27 Is the use of end-exhaled breath as a biological marker for TCE exposure for an environmental setting, a suitable method?

RQ28 What improvements are required to structure a feasible technique?