



Intrusion of Trichloroethylene (TCE) Vapors From Groundwater Into Buildings in Northeast MA

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Citation	Sidorenko, Irina. 2019. Intrusion of Trichloroethylene (TCE) Vapors From Groundwater Into Buildings in Northeast MA. Master's thesis, Harvard Extension School.
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Intrusion of Trichloroethylene (TCE) Vapors from Groundwater
into Buildings in Northeast MA

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A Thesis in the Field of Sustainability and Environmental Management
for the Degree of Master of Liberal Arts in Extension Studies

Harvard University

November 2018

Abstract

People can be exposed to trichloroethylene (TCE) in homes and businesses from vapors that intrude into buildings from subsurface groundwater and soil. Estimation of health risks is based on measured or modeled vapor concentrations in these buildings. The United States Environmental Protection Agency (U.S. EPA) recommends the use of empirically derived vapor attenuation factors for risk-based screening of the vapor intrusion pathway. The use of these recommended attenuation factors is expected to overestimate indoor air TCE concentrations.

This thesis research examined attenuation factors empirically derived from publicly available hazardous waste site data and identified key predictors and uncertainties. The primary objective of this research was to evaluate whether groundwater is an adequate predictor of indoor air exposure, focusing primarily on Trichloroethylene (TCE), a known human carcinogen and developmental toxicant. The primary research question was: Are current recommended U.S. EPA and MassDEP attenuation factors for TCE reliable for predicting indoor air concentrations of TCE? I answered this question by testing the following hypothesis: recommended attenuation factors for TCE are adequate predictors of indoor air concentrations of TCE for the protection of human health.

This study used data from 18 buildings that are hydraulically downgradient properties of TCE-contaminated sites, with distances from the building not exceeding 30 feet, where depth to groundwater was not more than 25 feet, and where there was no

reported soil source contamination. Attenuation factors were calculated for 18 buildings with paired indoor air and groundwater data and for 10 buildings with paired indoor air and subsurface soil gas data for TCE.

For five out of 18 buildings, the mean attenuation factor value between groundwater and indoor air was slightly higher, on average by less than one order of magnitude, than the empirically derived values recommended by U.S. EPA. For five out of 18 buildings, the mean attenuation factor value was higher by one order of magnitude than the value used by Massachusetts Department of Environmental Protection (MassDEP) in its calculation of the regulatory GW-2 standard. In both cases, TCE indoor air concentrations in some buildings were under-predicted. For five out of 10 buildings, the mean attenuation factor between subsurface soil gas and indoor air was higher on average by one order of magnitude than the empirically derived values recommended by U.S. EPA. For four out of 10 buildings, the mean attenuation factor was higher on average by half order of magnitude than recommended by MassDEP. In both cases, TCE indoor air concentrations in some buildings were under-predicted.

The results show that reliance on recommended U.S. EPA empirically derived attenuation factors and the vapor attenuation value used by MassDEP in its calculation of the regulatory GW-2 standard for predicting indoor air concentrations of TCE could under-predict vapor intrusion risk and may not protect health in some circumstances.

Dedication

I dedicate this thesis to my mother, who laid a firm foundation for my lifelong willingness to learn, who invested so much in my education and upbringing. You are the person I admire and I owe you for my academic and career success.

Acknowledgements

I wish to express my appreciation and gratitude for the people who have contributed to this project and remained supportive through the process. I am particularly thankful to Dr. Andy Friedmann and the vapor intrusion team from MassDEP, who introduced me to the world of vapor intrusion and inspired my true potential to take on this research. I am deeply grateful to my thesis director Dr. Wendy Heiger-Bernays who agreed to guide me through this challenging thesis writing process. Dr. Heiger-Bernays has been very supportive and her professional insights, expertise and concern for the problem guided the development of my own critical thinking about the subject and strengthened my analysis and writing skills.

I am deeply grateful to my partner Alex Mahilnitski, whom I met during my thesis writing process and who despite being far away in London, UK most of the time, was able to immensely support me during thesis development ups and downs. His endless solicitude and encouragement helped me cope with this personal challenge.

I want to express gratitude to my thesis proposal classmates and Professor Mark Leighton. Dr. Leighton helped me a lot with the initial stage of my research and provided a firm foundation of scientific research best practices. For my fellow classmates – I am so proud of all your thesis endeavors! Nikki Kocher – thank you for your positive attitude and enthusiasm, the fact that you were able to successfully complete your thesis despite many obstacles made me realize that everything is possible.

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Definition of Terms

310 CMR 40: Code of Massachusetts Regulation Title 310, a compilation of Department of Environmental Protection regulations.

Contaminated Groundwater: Groundwater containing oil and/or hazardous material at concentrations equal to or greater than a release notification threshold established by 310 CMR (definition is taken from 310 CMR 40).

Downgradient: (a) in reference to surface water, the direction perpendicular to lines of equal elevation over a distance in which elevation continuously decreases, measured from the point or area in question; or (b) in reference to groundwater, the direction perpendicular to lines of equipotential over a distance in which total head continuously decreases, measured from the point or area in question (definition is taken from 310 CMR 40).

Downgradient Property: A parcel of land which is located downgradient of the parcel of land which is the source of a release which has come to be located thereon (definition is taken from 310 CMR 40).

Groundwater: Any water below the earth's surface in the zone of saturation (definition is taken from 310 CMR 40).

GW-2 standard: Category of MCP standard for groundwater where there is a potential for volatile material to migrate into indoor air.

MassDEP: Massachusetts Department of Environmental Protection.

MCP: Massachusetts Contingency Plan and MCP each means 310 CMR 40.0000 (definition is taken from 310 CMR 40).

M.G.L. Chapter 21E: Massachusetts Oil and Hazardous Material Release Prevention and Response Act.

RC: Reportable concentration, concentration of oil or hazardous material in soil or groundwater which requires notification to the Department under M.G.L. c. 21E, and/or 310 CMR (definition is taken from 310 CMR 40).

Route of Exposure: A mechanism by which an oil or hazardous material comes into contact with a receptor, including, but not limited to, ingestion, inhalation, dermal absorption and transpiration (definition is taken from 310 CMR 40).

Site: A place or area from or at which a release of oil and/or hazardous material has occurred or where a threat of release exists (definition is taken from 310 CMR 40). Site can include multiple buildings.

Subslab depressurization (SSD) systems: An SSD system is designed to create a negative pressure below the building slab to prevent vapors from entering the building.

TCE: Trichloroethylene.

TVr: Residential indoor air threshold.

TVc/i: Commercial/industrial indoor air threshold.

U.S. EPA: United States Environmental Protection Agency.

Volatilization: Conversion of all or part of a liquid or solid into vapor (definition is taken from 310 CMR 40).

Volatile Organic Compounds (VOCs): Organic chemicals that have a high vapor pressure and can aggressively move by diffusion in air from groundwater and/or soil into adjacent environmental media.

Chapter I

Introduction

This thesis is structured in format for ready submission to a scientific journal, so that Chapter II is a draft of a journal article. Hence some of the background information as well as materials and methods are presented in Chapter II to avoid redundancy. In Chapter I, an overview of the research is given with its main questions and hypotheses, along with background information that is additional to the article. Chapter III is focused on discussion and conclusions.

This research is concerned with vapor intrusion (VI), a process by which vapor-forming chemicals migrate from any subsurface source into an overlying building (U.S. EPA, n.d.-1). It uses empirically derived data from publicly available sources and evaluates current State and Federal recommendations on vapor intrusion.

Vapor intrusion regulations and guidance documents exist on both Massachusetts State and Federal levels. In 1993, the Massachusetts Department of Environmental Protection (MassDEP) established threshold value standards for use at sites contaminated by releases of oil/hazardous materials. These standards were updated several times, most recently in 2014 (MassDEP, 2014-c, MassDEP, 2014-d). The most recent version of the MassDEP Vapor Intrusion Guidance was released in 2016 (MassDEP, 2016-b) and is intended to assist involved parties in complying with regulatory standards of

Massachusetts General Law chapter 21E and the Massachusetts Contingency Plan (the MCP or 310 CMR 40).

Research Significance and Objectives

To date, there has been a paucity of analyses to validate the underlying technical basis of existing Massachusetts vapor intrusion regulations. Fitzpatrick and Fitzgerald (2002) used field site data from 1996 to evaluate whether threshold groundwater concentrations of Volatile Organic Compounds (VOCs) are predictive of indoor-air exposure. Their results suggested that current standards are not always protective of public health (Fitzpatrick & Fitzgerald, 2002). My research addresses this need to evaluate the evidence for the most current vapor intrusion threshold values.

Therefore, the primary objective of this research is to evaluate whether groundwater is an adequate predictor of indoor air exposure, focusing primarily on Trichloroethylene (TCE), a known human carcinogen and developmental toxicant. This study addresses broader implications of state and federal regulations and the current reliance on groundwater as a predictor of indoor air for VOCs.

Background

Migration of chemical vapors from groundwater and/or soil into overlying buildings can cause a long-term hazard to human health. As the average American spends over 21 hours per day indoors and roughly 18 hours indoors for every hour spent outdoors (Olson & Corsi, 2002), the potential presence of harmful vapors in buildings is of great importance. Until the 1990s, most work in the area of vapor intrusion research

was related to radon, whose radioactive nature was the basis of health concerns for individuals residing in impacted structures (Bozkurt, 2009). More recently the focus has broadened towards other toxic chemicals of a volatile nature. The current research is focused on vapor intrusion of one chemical, trichloroethylene (TCE), which the U.S. EPA has classified as a human carcinogen and a developmental toxicant regardless of the route of exposure (U.S. EPA, n.d.-2).

TCE Toxicity and Regulation

Trichloroethylene falls into the category of Volatile Organic Compounds (VOCs) – chemicals that have a high vapor pressure and can aggressively evaporate from contaminated groundwater and/or soil as it has a vapor pressure of 74 mmHg (U.S. EPA, 2016). According to the Agency for Toxic Substances and Disease Registry, trichloroethylene is the most frequently reported organic contaminant in groundwater (ATSDR, 2014). The chemical can be released into the environment during the course of manufacture and formulation, and during its use. TCE is primarily used as a degreaser in the metal and automotive industry. It is also used in some dry cleaning agents and in some consumer products such as paint removers, gun cleaners, glues and spray fixatives for arts and craft uses (ATSDR, 2014).

Trichloroethylene is a potent carcinogen and is linked to various harmful human health effects (Chiu et al., 2013) including cardiac malformations in the fetus (U.S. EPA IRIS, 2011; U.S. EPA, 2011). The health impact of inhaling TCE is dependent on its concentration in indoor air, length of exposure and on individual variables such as whether a pregnant woman is exposed. Health outcomes of TCE exposure are discussed in

Chapter II. In order to protect public health, MassDEP has developed TCE threshold values for groundwater, indoor air and subsurface soil gas – vapors in the air spaces between soil particles (also called sub slab soil gas or just sub slab).

Current Massachusetts Regulations

In 1993, the Massachusetts Department of Environmental Protection (MassDEP) established generic groundwater cleanup standards for use at sites contaminated by releases of oil/hazardous materials (Fitzpatrick & Fitzgerald, 2002). Where potential exists for migration of volatile chemicals into indoor air, a GW-2 category is assigned for groundwater (MassDEP, 2014-c). MassDEP's Groundwater Category GW-2 standard is applied to groundwater near occupied buildings and designed to be protective of indoor air (MassDEP, 2010). According to Massachusetts Contingency Plan (MCP), GW-2 groundwater must be within 30 feet of a building and the average annual depth to groundwater must be 15 feet or less (MassDEP, 2014-c). In 2014, MassDEP updated the TCE GW-2 standards for groundwater and threshold values for indoor air at residential and commercial buildings. New information about the potential toxicity of TCE has resulted in MassDEP developing more stringent screening levels for TCE (Table 1).

Table 1. TCE GW-2 standards for groundwater and threshold values for indoor air in the Commonwealth of Massachusetts (MassDEP, 2014-c).

	Groundwater	Indoor Air	Exposure Duration	Exposure Frequency
Residential	5 µg/L	0.4 (µg/m ³)	24 hours per day	365 days per year
Commercial	5 µg/L	1.8 (µg/m ³)	8 hours per day	250 days per year

Vapor Attenuation Factor

Vapor attenuation is the reduction in concentration of VOCs that occurs during vapor migration in the subsurface (U.S. EPA, 2012). Attenuation factors are often used when assessing contaminated sites; these factors permit estimation of contaminant indoor air concentrations in the structure of concern by relating these to corresponding groundwater and subsurface soil gas concentrations. In general, vapor intrusion guidance can vary from state to state and can be different from federal recommendations; nevertheless, EPA has been working towards providing national guidance on the issue. Massachusetts DEP and Federal US EPA recommended attenuation factors are shown in Table 2.

Table 2. Recommended U.S. EPA and MassDEP Attenuation Factors for TCE.

	MassDEP (MassDEP, 2017)		U.S. EPA (U.S. EPA, 2015)	
	Groundwater to Indoor Air	Soil Gas to Indoor Air	Groundwater to Indoor Air	Soil Gas to Indoor Air
Scientific Notation	7.53E-04	3E-02	1E-03	1.4E-02
Decimal Form	0.000753	0.03	0.001	0.014

To calculate an attenuation factor from empirical data, the indoor air concentration of the chemical is divided by its subsurface concentration. When the attenuation factor is larger there is less chemical volatilization and more vapor migration from subsurface source to indoor air in an overlying building; therefore, large attenuation factors result in higher concentrations indoors. A smaller attenuation factor infers more chemical volatilization and less vapor migration from subsurface source to indoor air in

an overlying building and lower indoor air concentration. In general, more attenuation means more volatilization and less vapor migration, hence lower indoor air concentration and vice versa - less attenuation means more vapor migration and less volatilization, therefore higher indoor air concentration.

Validation of Existing Regulations

Vapor intrusion is an area of active research, but there is a scarcity of peer-reviewed articles that use field data from sites where the vapor intrusion pathway of VOCs exists. And even fewer studies exist that focus on evaluating existing regulations and guidance on vapor intrusion using empirically derived site data. The most current attempt to validate existing Massachusetts State regulations was done in 1996 by Fitzpatrick and Fitzgerald (2002). The researchers used field data from 47 contaminated sites where VOCs had impacted groundwater and/or indoor air, with 11 of these being TCE sites (Fitzpatrick & Fitzgerald, 2002). Attenuation factors were calculated between sub-slab soil gas and indoor air, but a comparison between groundwater and indoor air was not conducted. There were other limitations in the study. The researchers did not account for data variability, using only one combination of samples for each site. Moreover, the study failed to detail the methodology used to combine pairs of samples. Finally, professional judgment was used to reconcile anomalies or data gaps, which together with the methodological ambiguities described above can lead to biased and unreproducible results.

The U.S. EPA has provided national guidance on vapor intrusion (U.S. EPA, 2016), but the guidance is not generally agreed upon by environmental scientists and risk

assessors (Bozkurt, 2009). Regulatory guidelines and standards for levels of TCE concentration in groundwater, soil gas and indoor air vary from one state to another and different state guidance arguably demonstrates limited understanding of this complex issue (Bozkurt, 2009). Interpretation of field data and the assessment of vapor intrusion pathways are widely recognized as challenging, because TCE concentrations in adjacent structures can vary dramatically and inconsistencies are very common due to multi-factor variability (Fitzpatrick & Fitzgerald, 2002). Therefore, variability in data must be taken into account in order to assess the volatile behavior of TCE and its potential intrusion into buildings.

Research Question, Hypothesis and Specific Aims

To help address these shortcomings, my primary research question was: Are current recommended U.S. EPA and MassDEP attenuation factors for TCE reliable for predicting indoor air concentrations of TCE? I answered this question by testing the following hypothesis: recommended attenuation factors for TCE are adequate predictors of indoor air concentrations of TCE for the protection of human health.

I evaluated a range of empirically derived attenuation factors, their numerical distribution and compared the values with default or recommended U.S. EPA and MassDEP attenuation factors with the aim of determining whether the default values adequately model the environment. I hypothesized that the majority of attenuation factors are within the range of recommended values.

Finally, I explored the relationship between attenuation of TCE vapors between groundwater and indoor air by performing linear regression analysis between maximum

TCE concentrations in indoor air and maximum TCE groundwater concentrations across buildings in the study using the empirical data extracted from reports submitted to MassDEP. I hypothesized that a positive but weak correlation between groundwater and indoor air concentrations would be found.

Specific Aims

The key aims of this research were to:

1. Create a database of TCE contaminated hazardous waste sites in the Northeast region of Massachusetts based on defined inclusionary criteria.
2. Pair groundwater and indoor air data samples, as well as subsurface soil gas and indoor air data (where available) using a defined methodology.
3. Calculate attenuation factors for paired samples.
4. Perform statistical analyses of the calculated vapor attenuation factors.
5. Compare state and federal recommendations with results derived from the empirical data.
6. Determine whether the findings support my hypotheses.

Methods are described in the second chapter of this thesis.

Chapter II

Data-driven Determination of Attenuation Factors for Prediction of Trichloroethylene Vapor Intrusion into Buildings in Northeast MA

Abstract

People can be exposed to trichloroethylene (TCE) in homes and businesses from vapors that intrude into buildings from subsurface groundwater and soil. Estimation of health risks is based on measured or modeled vapor concentrations into these buildings. The United States Environmental Protection Agency (U.S. EPA) recommends the use of empirically derived vapor attenuation factors for risk-based screening of the vapor intrusion pathway. The use of these recommended attenuation factors is expected to overestimate indoor air TCE concentrations.

This research examined attenuation factors empirically derived from publicly available hazardous waste site data and identified key predictors and uncertainties. This study used data from 18 buildings that are hydraulically downgradient properties of TCE-contaminated sites, with distances from the building not exceeding 30 feet, where depth to groundwater was not more than 25 feet, and where there was no reported soil source contamination. Attenuation factors were calculated for 18 buildings with paired indoor air and groundwater data and for 10 buildings with paired indoor air and subsurface soil gas data for TCE. For five out of 18 buildings, the mean attenuation factor value between groundwater and indoor air was slightly higher, on average by less than one order of magnitude, than the empirically derived values recommended by U.S. EPA. For five out

of 18 buildings, the mean attenuation factor value was higher by one order of magnitude than the value used by Massachusetts Department of Environmental Protection (MassDEP) in its calculation of the regulatory GW-2 standard. In both cases, TCE indoor air concentrations in some buildings were under-predicted. For five out of 10 buildings, the mean attenuation factor between subsurface soil gas and indoor air was higher on average by one order of magnitude than the empirically derived values recommended by U.S. EPA. For four out of 10 buildings, the mean attenuation factor was higher on average by half order of magnitude than recommended by MassDEP. In both cases, TCE indoor air concentrations in some buildings were under-predicted.

The results show that reliance on recommended U.S. EPA empirically derived attenuation factors and the vapor attenuation value used by MassDEP in its calculation of the regulatory GW-2 standard for predicting indoor air concentrations of TCE could under predict vapor intrusion risk and may not be health protective in some circumstances.

Introduction

Trichloroethylene (TCE) is a chlorinated solvent that has a history of being used as a degreaser in the metal and automotive industries, in some dry cleaning agents and in some consumer products such as paint removers, gun cleaners, glues and spray fixatives for arts and crafts, and is the most frequently reported organic contaminant in groundwater (ATSDR, 2014). Products with TCE are not widely available in all US states, but a non-inclusive list of products can be found in the Household Products Database compiled by U.S. Department of Health & Human Services (U.S. NHS, n.d.).

Some products such as gun cleaner, which can be purchased online, is composed of nearly 90-99% TCE, according to the Occupational Safety and Health Administration (OSHA, 2016).

Epidemiological and toxicological evidence support the classification of TCE (CASRN 79-01-6) as a human carcinogen (U.S. EPA, 2011). Increased incidences of tumors in the kidney, liver and lymphoid tissues have been reported in rats and mice exposed to high levels of TCE via inhalation and oral exposure (Chiu et al., 2013). Human data provide strong support for TCE-induced kidney and liver cancer, malignant lymphoma in humans (Scott & Jinot, 2011) as well as evidence for associations between exposure to multiple VOCs, including TCE in contaminated drinking water and male breast cancer at U.S. Marine Corps Base Camp Lejeune (Ruckart et. al., 2015). In addition, TCE is linked to a number of non-cancer health outcomes (Chiu et al., 2013). Exposure to elevated concentrations of trichloroethylene vapors can result in central nervous system depression, loss of consciousness and even death (ATSDR, 2014). Available human and animal data identify the heart, kidney, liver, immune system, male reproductive system and a developing fetus as targets of trichloroethylene toxicity (ATSDR, 2014). Increased prevalence of cardiac defects was found in children with exposure of either parent during the first trimester of pregnancy to well water contaminated with TCE (Goldberg et al., 1990). Pregnant Sprague-Dawley rats treated daily with 500 mg/kg of TCE in drinking water demonstrated an increased incidence of cardiac malformations in developing rat fetuses (Johnson et al., 2003). Fetal cardiac malformation as an important outcome for human exposure and is a defensible endpoint

that is supported by in vivo, in vitro and molecular pathway studies (MassDEP, 2014-a, MassDEP, 2014-b).

The Massachusetts Department of Environmental Protection (MassDEP) (MassDEP, 2016) and United States Environmental Protection (U.S. EPA) (U.S. EPA, 2015-a) have developed guidance documents intended to provide practitioners with a roadmap to evaluation and elimination of exposure pathways to TCE indoors from vapor intrusion. Indoor sources of TCE are typically not the purview of regulatory programs, and so elimination of indoor uses is addressed through education and behavioral changes by people using the TCE-containing products. The human health impact of inhaling TCE is dependent on its concentration in indoor air, length of exposure and on individual variables such life stage. Since data support TCE as a developmental toxicant, pregnant women are considered susceptible populations.

In 1993, MassDEP established generic groundwater cleanup standards for use at sites contaminated by releases of oil/hazardous materials (Fitzpatrick & Fitzgerald, 2002). In this system, groundwater is categorized based on its use, location and discharge to surface water. Where potential exists for migration of volatile chemicals into indoor air, a GW-2 category is assigned to the groundwater (MassDEP, 2014-c). According to the Massachusetts Contingency Plan (MCP), GW-2 groundwater is within 30 feet of a building and the average annual depth to groundwater must be 15 feet or less (MassDEP, 2014-c). In 2014, MassDEP promulgated TCE threshold concentration values for groundwater and indoor air at residential and commercial buildings - 5 µg/L for residential and commercial groundwater (GW) concentrations and 0.4 µg/m³ for residential and 1.8 µg/m³ for commercial indoor air (IA) concentrations.

Vapor attenuation factor (AF) is the reduction in concentration of VOCs that occurs during vapor migration from a subsurface source into a building (U.S. EPA, 2012). When combined with an appropriate attenuation factor, groundwater data can be used to estimate a potential upper-bound indoor air concentration that may arise from vapor intrusion (U.S. EPA, 2015-a). The attenuation factor is calculated by dividing the indoor air concentration by the subsurface concentration, so when the attenuation factor is larger there is less chemical volatilization from subsurface source to indoor air in an overlying building; therefore, larger attenuation factors result in less attenuation and higher concentrations indoors (Figure 1).

The U.S. EPA compiled an empirical data set from 913 buildings and 41 sites and based on analyses recommended empirically derived attenuation factors (AF), also known as default or generic attenuation factors (U.S. EPA, 2015-a). Default attenuation factors are often used when assessing contaminated sites and the U.S. EPA has recommended value of $1\text{E-}03$ for groundwater (GW) and $3\text{E-}02$ for subsurface soil gas (SS) (U.S.EPA, 2015-a). The smaller the attenuation factor, the more chemical volatilization is taking place from subsurface source to indoor air in an overlying building, therefore smaller concentration indoors. The MassDEP GW-2 standards are intended to limit indoor air concentrations of vapors from the groundwater into indoor spaces. The GW-2 standards were derived using chemical-specific attenuation factors that are also used for preliminary vapor intrusion screening, for TCE attenuation factor of $7.53\text{E-}04$ is used and is listed in the MCP (MassDEP, 2017). For subsurface soil gas, MassDEP uses an attenuation factor of $1.4\text{E-}02$ to estimate subsurface soil gas to indoor air attenuation for TCE (MassDEP, 2016).

The attenuation of vapors from groundwater to indoor air can be highly variable due to spatial and temporal variability and can vary widely from site to site and from building to building within a site (Holton et al. 2013, F&F 2002, U.S. EPA 2012).

More attenuation means more chemical volatilization, more reduction, less vapor migration, lower indoor air concentration.

Less attenuation means less volatilization, less reduction, more vapor migration, higher indoor air concentration.

Larger AF – less volatilization, higher indoor air concentration.

Smaller AF – more volatilization, lower indoor air concentration.

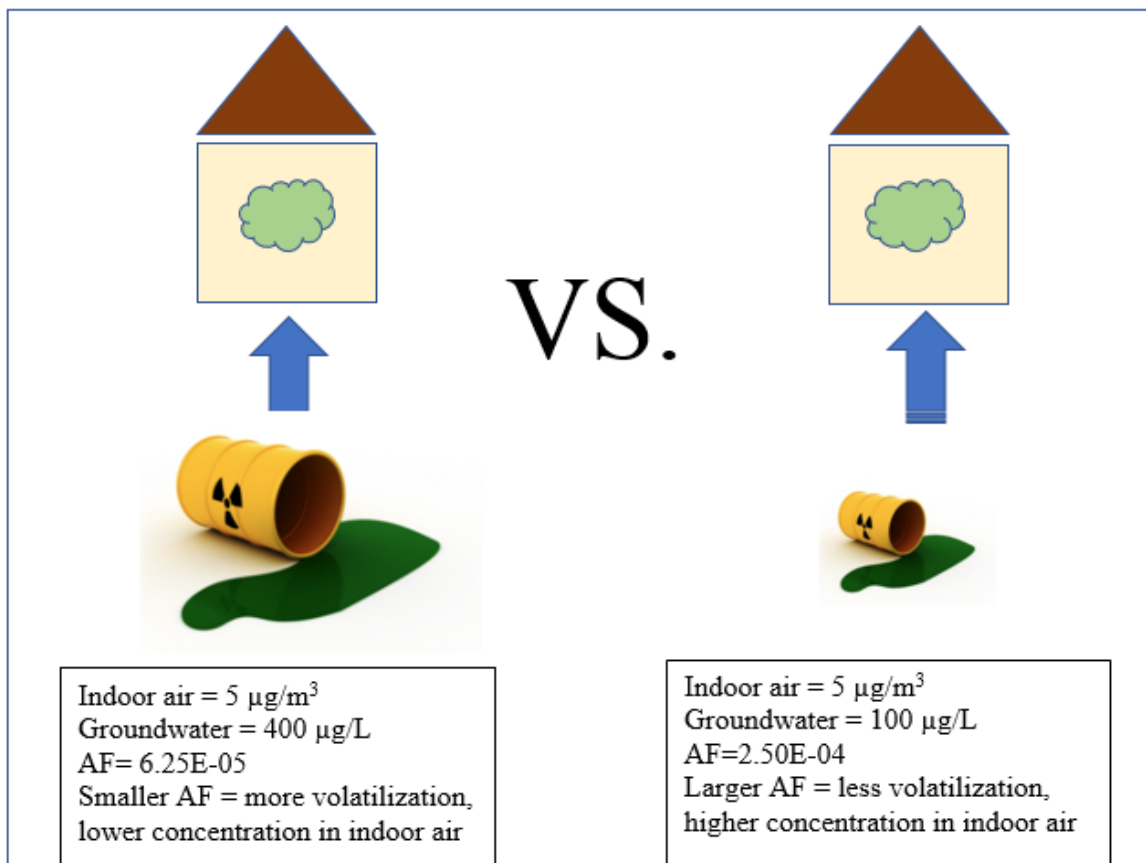


Figure 1. Small vs. large attenuation factor and interpretation of predicted concentrations in indoor air.

The variability in attenuation can be due to multiple factors, e.g., groundwater flow patterns, contaminant source distribution features, soil gas entry rates, air exchange, etc. Factors also can vary over time within a building depending on the time of the year, whether it's winter when windows are usually closed and heating systems are more active or summer with more air conditioning and ventilation. Due to this variability, it is useful to use empirically derived factors in order to determine the most important predictors of vapor intrusion. Therefore, this study was undertaken to determine relationships between subsurface sources of TCE and indoor air, where empirically derived attenuation factors from the database in this study are compared with default or recommended attenuation factors with the aim of determining whether the default values adequately model the environment.

Materials and Methods

Selection of Data from Publicly Accessible Portal

Groundwater, subsurface and indoor air TCE data from a subset of M.G.L. Chapter 21E Hazardous Waste Sites were extracted from the MassDEP public Searchable Sites Database (MassDEP, <https://eeaonline.eea.state.ma.us/portal#!/search/wastesite>) using the following inclusion criteria: 1) groundwater well distance from the building was not more than 30 feet; 2) sample depth to groundwater was not more than 25 feet, although due to temporal/seasonal variation, depth to groundwater varies and average sample depth was used; 3) sites were screened for soil contamination source to exclude the possibility of vapors from the contaminated soil source; and 4) sample and analysis reports contained information about building type, foundation type, soil characteristics,

other VOCs measured, whether indoor air samples were taken after removal background or in-building sources, if and when a Sub Slab Depressurization (SSD) System was installed. This information, as well as concentration measurements for TCE in groundwater, subsurface soil gas (if available) and indoor air were extracted and placed into an Excel database. Where concentrations for TCE were reported as non-detects in groundwater and subsurface soil gas, samples were excluded, only non-detects in indoor air were included. Non-detect values in indoor air were replaced with $\frac{1}{2}$ the reporting limit. The location of each indoor air sample was noted in the database, e.g., basement, 1st/2nd floor and using report site plans, the closest groundwater well was indicated for each indoor air sample to help pair the samples for calculating attenuation factors.

For this analysis, definition of site corresponds to how MCP defines it and is used to refer to a place or area from or at which a release of oil and/or hazardous material has occurred or where a threat of release exists (MassDEP, 2017). Eleven sites were selected from more than 1,000 reviewed reports where a release of TCE was evaluated for impact to indoor air. Some sites had multiple buildings that were affected by the release and a total of 18 buildings were included in the database. All buildings were hydraulically downgradient to the MassDEP-identified contamination source, meaning contaminated groundwater flowed in the direction of the steepest gradient.

Calculating Attenuation Factors

All indoor air samples were paired with groundwater samples. In order to account for the uncertainty of which the groundwater sample was responsible for particular indoor air concentrations, each groundwater sample concentration was paired with each

available indoor air sample. By example, as shown in Table 3, the two groundwater samples were paired with the two indoor air samples, resulting in four attenuation factors. Given that all evaluated data was extracted from environmental reports that were produced by others, in most cases there was a time gap between taking groundwater, subsurface soil gas and indoor air samples, ranging from being weeks to over a year apart. Therefore, the pairing approach that was used in the study accounts for the fact that any of the groundwater samples could potentially contribute to any of the indoor air concentrations. The same logic was used when pairing subsurface soil gas and indoor air samples. Where there are groundwater and subsurface soil gas samples available in a building, groundwater samples were paired with indoor air samples and subsurface soil gas samples were paired with the same available indoor air samples.

Table 3. Pairing groundwater and indoor air samples for calculating vapor attenuation factors.

GW sample	TCE in GW	$[TCE_{gw}] \times H \times 1000 \text{ (L/m}^3\text{)}$	TCE in IA	Vapor Attenuation Factor
	$\mu\text{g/L}$	$\mu\text{g/m}^3$	$\mu\text{g/m}^3$	unitless
1	120	24000	3.5	1.46E-04
1	120	24000	1.5	6.25E-05
2	89	17800	3.5	1.97E-04
2	89	17800	1.5	8.43E-05

Attenuation factors were calculated for 18 buildings with paired indoor air and groundwater TCE data and for 10 buildings with paired indoor air and subsurface soil gas TCE data. As a result, 84 groundwater samples were paired with 99 indoor air samples, and 55 subsurface soil gas samples with 72 indoor air samples. All paired samples were taken within a two-year span (with the exception of two groundwater samples that are

noted in the database). When indoor air data was available on several levels of the buildings, only lowest level data (basement or 1st floor) was used for pairing.

Consistent with the state of the practice, groundwater concentrations (C_{GW}) were converted into vapor concentrations assuming equilibrium conditions (i.e., by multiplying the groundwater concentration by the chemical's dimensionless Henry's Law constant at the specified groundwater temperature) (U.S. EPA, 2012). Attenuation factors were calculated using the following equation:

$$AF_{VI} = \frac{C_{IA}}{C_{SV}} = \frac{C_{IA}}{C_{GW} * H'_{TS}}$$

where AF_{VI} is the vapor attenuation factor (unitless); C_{IA} is the vapor concentration at the source of contamination ($\mu\text{g}/\text{m}^3$); C_{SV} = subsurface source vapor TCE concentration ($\mu\text{g}/\text{m}^3$), In the case of groundwater as the vapor source, C_{sv} is estimated assuming that the vapor and aqueous phases are in local equilibrium according to Henry's law; H'_{TS} = Henry's law constant at the system (groundwater) temperature (dimensionless) (for TCE= 0.2); C_{GW} = concentration of volatile substance in groundwater ($\mu\text{g}/\text{L}$).

Statistical Analyses of the Calculated Vapor Attenuation Factors

To evaluate overall attenuation factors' range and distribution and to compare the values to recommended U.S. EPA and MassDEP attenuation factors, a frequency distribution analysis was conducted for all groundwater to indoor air and subsurface soil gas to indoor air attenuation factors. Histograms were created in Excel to present the distribution of all attenuation factors across 18 buildings.

Box-and-whisker plots were generated for groundwater to indoor air and for subsurface soil gas to indoor air attenuation factors for each building in the study to present the distribution of attenuation factors on a building level. The attenuation factors were compared with recommended U.S. EPA and MassDEP attenuation factors.

Linear regression was performed in order to test relationship between maximum TCE concentration in indoor air and maximum TCE groundwater concentration across buildings in the study. The statistical software package, ProUCL was used to calculate 95% upper confidence limit (UCL) for each building and used for regression analysis (U.S EPA, 2013).

Multiplier of GW-2 standard and indoor air threshold: for each building, the maximum TCE concentration in groundwater was divided by the MassDEP GW-2 standard for TCE (5 $\mu\text{g/L}$). For indoor air, the maximum TCE concentration in indoor air was divided by the residential threshold (TVr) of 0.4 $\mu\text{g/m}^3$ or commercial threshold (TVc/i) of 1.8 $\mu\text{g/m}^3$ for TCE in indoor air as set by MassDEP (MassDEP, 2016). Results were graphed to examine the magnitude of deviation from the GW-2 standard and indoor air threshold for groundwater and indoor air. Percentage differences were calculated and included in the graph.

Results

The range, mean, median and standard deviation values for all groundwater to indoor air and subsurface soil gas to indoor air attenuation factors are presented in Table 4. The same measures for each of the 18 buildings are shown in Tables 5 and 6. Table 5 presents groundwater to indoor air and Table 6 presents subsurface soil gas to indoor air

range, mean, median and standard deviation values for calculated attenuation factors.

Since attenuation is the ratio of indoor air concentration to subsurface concentration (groundwater or subsurface soil gas) and is used as a measure of the decrease in concentration that occurs during vapor migration, smaller attenuation factors therefore mean greater chemical volatilization and lower concentrations in the indoor air. Likewise, larger attenuation factors indicate that less attenuation and less volatilization is expected as VOC vapors migrate into a building, hence higher indoor air concentration.

Attenuation factors tend to be less than “1” since indoor air TCE concentrations at most times are lower than subsurface values. Recommended attenuation factors are intentionally conservative and protective, i.e., such that multiplying the concentration of a subsurface volatile organic compound (VOC) by the appropriate AF will overestimate the indoor concentration most of the time, and underestimate it only occasionally (Schmidt, 2014).

Table 4. Descriptive statistics for calculated attenuation factors from paired groundwater and subsurface TCE concentrations in all 18 buildings.

	GW to IA AFs	SS to IA AFs
Min	2.46E-08	3.07E-06
Max	7.80E-01	3.18E+00
Mean	2.41E-03	4.07E-02
Median	9.69E-05	1.77E-03
Standard Deviation	3.44E-02	1.69E-01

Frequency Distribution Analyses

A frequency distribution of groundwater to indoor air attenuation factors presents the range of 2.46E-08 - 7.80E-01 values (Figure 1). For groundwater to indoor air, 81

attenuation factors of 661 were above the recommended $7.53\text{E-}04$ value assumed by MassDEP in its calculation of the regulatory GW-2 standard, and 62 out of 661 were above the recommended U.S. EPA attenuation factor of $1\text{E-}03$, which means less attenuation and less volatilization between identified pairs and higher concentrations indoors.

A frequency distribution of subsurface soil gas to indoor air attenuation factors shows the range of $3.07\text{E-}06$ - $3.18\text{E+}00$ values (Figure 3). For subsurface soil gas to indoor air, 141 attenuation factors of 531 were above the recommended MassDEP attenuation factor of $1.4\text{E-}02$ and 107 attenuation factors out of 531 were above the recommended U.S. EPA attenuation factor of $3\text{E-}02$ which means less attenuation and less volatilization between identified pairs and higher concentrations indoors.

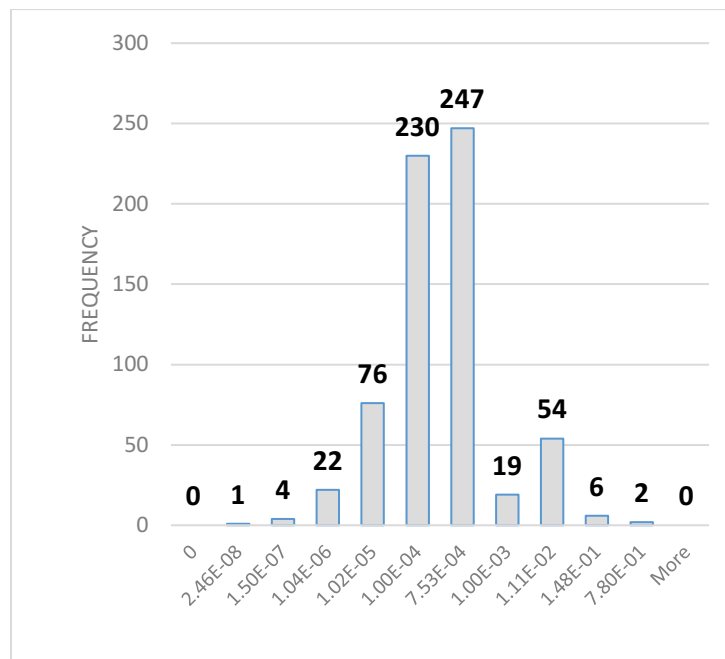


Figure 2. Groundwater to indoor air AF distribution across all (18) buildings.

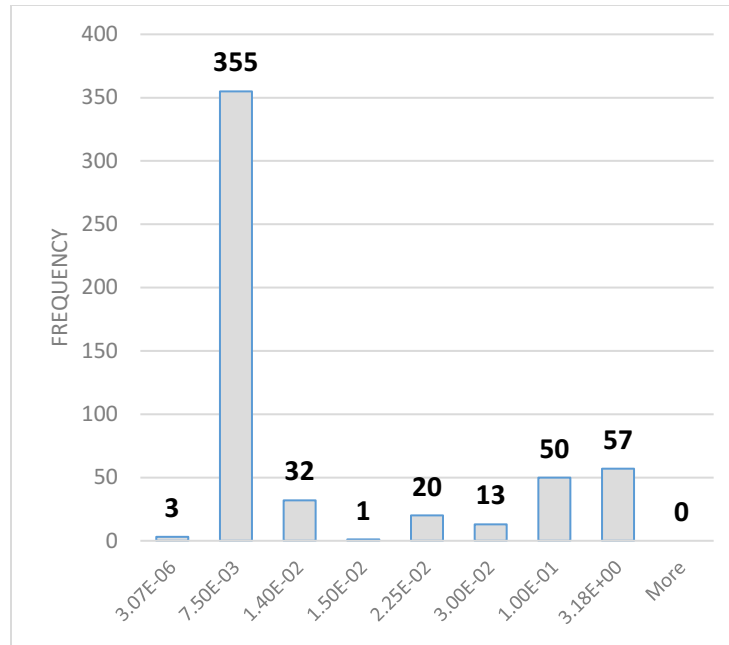


Figure 3. Subsurface soil gas to indoor air AF distribution across all (10) buildings.

The median value of all groundwater to indoor air attenuation factors across all buildings was $9.69\text{E-}05$ and the default AF of $1\text{E-}03$ recommended by U.S. EPA coincides with the 90th percentile data in this study dataset, making the AF sufficiently conservative to protect occupants in 90% of buildings. The default AF of $7.53\text{E-}04$ recommended by MassDEP coincides with the 87th percentile data, making the AF sufficiently conservative to protect occupants in 87% of buildings.

The median value of all subsurface soil gas to indoor air attenuation factors across all buildings was $1.77\text{E-}03$, and the default AF of $3.0\text{E-}02$ recommended by U.S. EPA coincides with the 80th percentile data, making the AF sufficiently conservative to protect occupants in 80% of buildings. The default AF of $1.40\text{E-}02$ recommended by MassDEP coincides with the 74th percentile data, making the AF sufficiently conservative to protect occupants in 74% of buildings.

On a building level, for five out of 18 buildings, the mean attenuation factor between groundwater and indoor air was slightly higher, on average less than one order of magnitude, than the empirically derived values recommended by U.S. EPA. For five out of 18 buildings, the mean attenuation factor was higher by one order of magnitude than the value used by MassDEP in its calculation of the regulatory GW-2 standard. In both cases, TCE indoor air concentrations in five buildings were under-predicted. Table 5 shows the attenuation factors distribution by building and Figure 4 shows box-and-whisker plots for each building summarizing the attenuation factor distribution for groundwater to indoor air.

For five out of 10 buildings, the mean attenuation factor value between subsurface soil gas and indoor air is higher on average by half order of magnitude than that recommended by U.S. EPA. For four out of 10 buildings, the mean attenuation factor is higher by average of half order of magnitude than recommended MassDEP value of $1.40\text{E-}02$, under-predicting TCE concentrations in indoor air in some cases. Table 6 shows the distribution in attenuation factors by building and Figure 5 shows box-and-whisker plots for each building.

Linear regression analyses of paired 95% UCL of maximum TCE indoor air concentrations and 95% UCL of maximum TCE groundwater concentrations on a building level shows a lack of association between groundwater and indoor air with an $R^2=0.0002$, meaning increase/decrease in groundwater concentration is not correlated with increase/decrease in indoor air concentrations of TCE.

Table 5. Descriptive statistics for calculated attenuation factors distribution from paired groundwater to indoor air for each of 18 buildings.

Building	18 Osprey	85 River	80 Fountain	100-120 Fountain	780 Beacon	304 Boston Post	311 Boston Post	10 Sun	8 Sun	17 Fern	57 Putnam	114 Dudley	2377 Washington	258 Beacon	3 Beckwith	148 Maple	320 Nevada	459 Watertown
	AF	AF	AF	AF	AF	AF	AF	AF	AF	AF	AF	AF	AF	AF	AF	AF	AF	AF
Min	2.62E-05	7.62E-06	9.42E-07	3.21E-06	2.46E-06	5.63E-04	2.81E-06	4.69E-08	7.54E-07	2.46E-08	3.79E-07	1.79E-06	6.25E-05	4.41E-05	1.86E-06	9.00E-06	2.74E-03	1.95E-03
Max	6.74E-03	3.72E-03	6.05E-04	2.61E-03	1.18E-05	4.02E-03	1.59E-05	3.68E-06	1.98E-06	1.58E-06	8.37E-05	2.44E-03	1.97E-04	7.80E-01	3.58E-04	1.73E-04	1.30E-02	2.32E-03
Mean	1.08E-03	5.21E-04	1.13E-04	3.56E-04	4.74E-06	1.77E-03	7.24E-06	7.94E-07	1.44E-06	3.77E-07	1.57E-05	4.23E-04	1.22E-04	5.76E-02	2.30E-04	7.94E-05	6.12E-03	2.10E-03
Median	1.48E-04	2.33E-04	9.07E-05	2.36E-04	3.48E-06	1.35E-03	5.47E-06	4.29E-07	1.59E-06	1.98E-07	1.09E-05	1.26E-04	1.15E-04	2.29E-03	3.31E-04	6.86E-05	5.63E-03	2.03E-03
Standard Deviation	2.18E-03	7.60E-04	1.06E-04	4.23E-04	3.50E-06	1.29E-03	4.95E-06	9.64E-07	6.28E-07	5.97E-07	1.77E-05	6.06E-04	6.08E-05	1.75E-01	1.98E-04	6.97E-05	3.71E-03	1.98E-04

Table 6. Descriptive statistics for calculated attenuation factors distribution from paired subsurface soil gas to indoor air for each of 10 buildings.

Building	85 River	80 Fountain	100-120 Fountain	780 Beacon	8 Sun	57 Putnam	114 Dudley	2377 Washingt	148 Maple	320 Nevada
	AF	AF	AF	AF	AF	AF	AF	AF	AF	AF
Min	1.56E-05	7.04E-05	1.58E-05	1.85E-02	1.16E-03	3.07E-06	2.94E-03	8.38E-03	4.43E-01	1.23E-03
Max	5.53E-01	5.60E-02	2.05E-02	1.85E-02	1.47E-02	1.45E-04	2.88E-01	3.18E+00	6.23E-01	3.27E-01
Mean	3.17E-02	5.07E-03	1.93E-03	1.85E-02	6.45E-03	2.60E-05	1.02E-01	7.03E-01	5.33E-01	8.32E-02
Median	2.38E-03	1.04E-03	1.05E-03	1.85E-02	4.32E-03	1.53E-05	7.57E-02	1.65E-01	5.33E-01	1.35E-02
Standard Deviation	7.92E-02	1.04E-02	3.05E-03	n/a	5.53E-03	2.77E-05	8.61E-02	1.11E+00	1.28E-01	1.07E-01

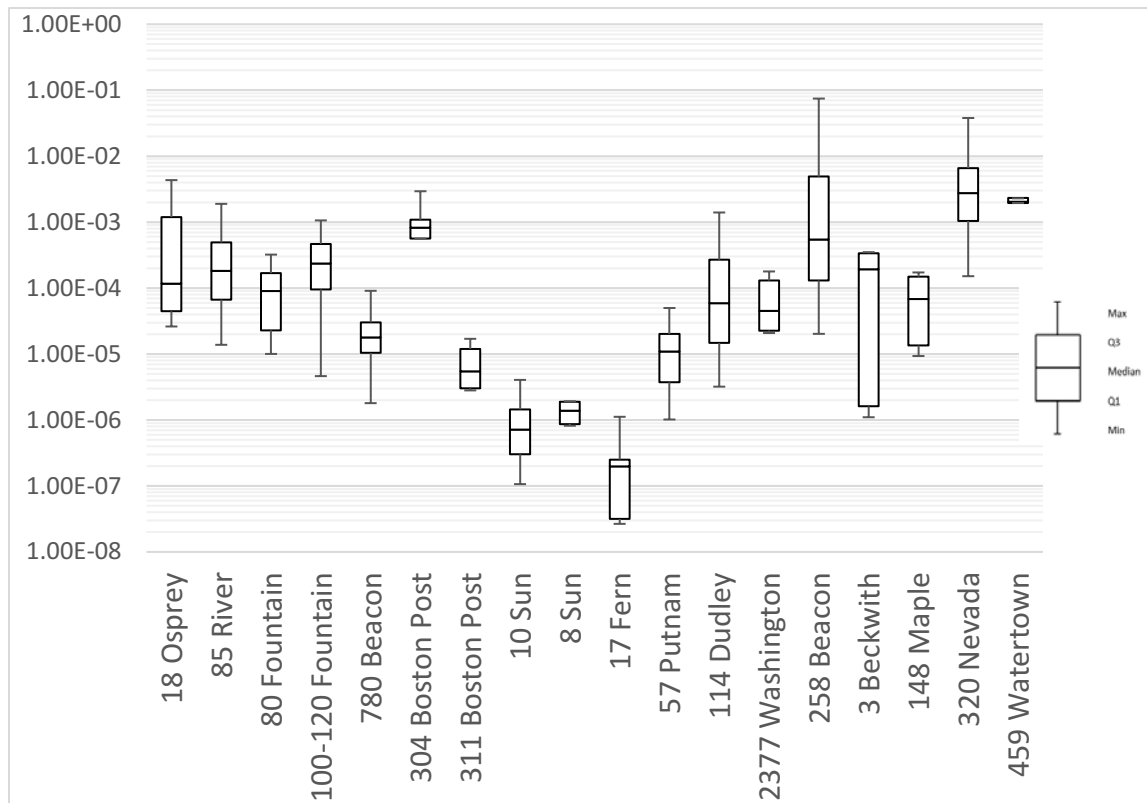


Figure 4. Box-and-whisker plot summarizing GW-IA attenuation factor distribution on a log scale (base 10) for each of the 18 buildings.

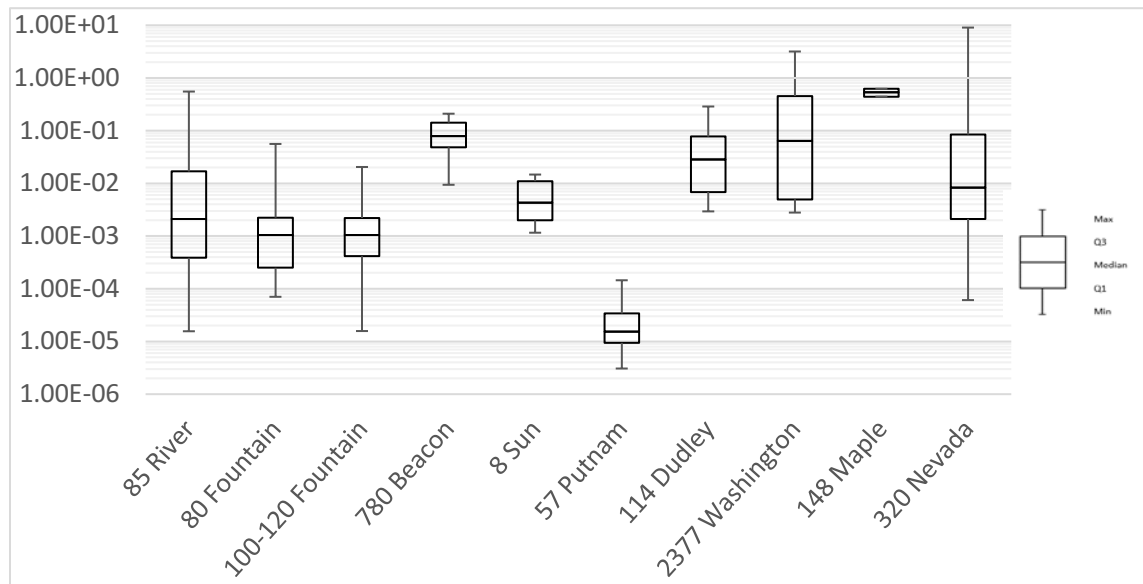


Figure 5. Box-and-whisker plot summarizing SS-IA attenuation factor distribution on a log scale (base 10) for each of the 10 buildings.

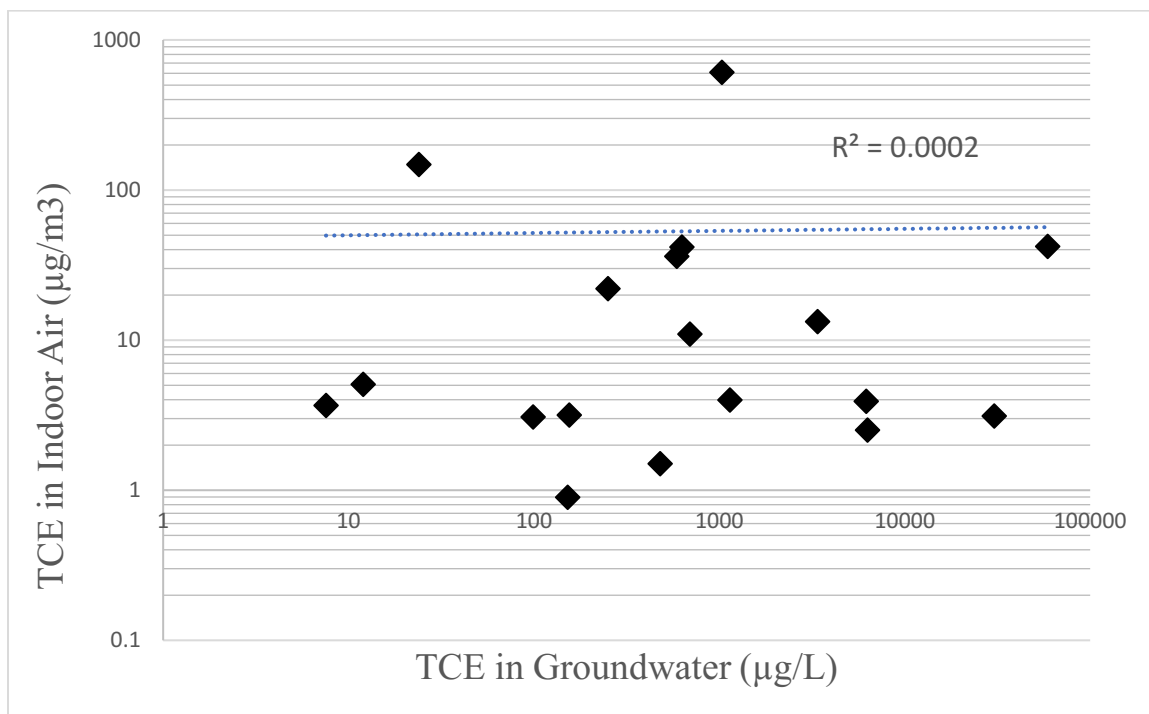


Figure 6. Linear regression of 95% UCL of maximum TCE IA ~ 95% UCL of maximum TCE GW on a log scale (base 10).

Multiplier of GW-2 standard and indoor air threshold value: Figure 6 shows the magnitude of deviation from the MassDEP GW-2 standard and indoor air threshold for maximum groundwater and indoor air concentrations on an individual building basis. Results indicate that greater deviation from the GW-2 standard for groundwater (meaning higher chemical concentration) does not result in higher deviation from TVr and TVv/i for indoor air ($r^2=0.00017$, n.s.). Nine buildings had about 200% percent difference between applicable regulatory standard deviation of groundwater and corresponding indoor air concentration.

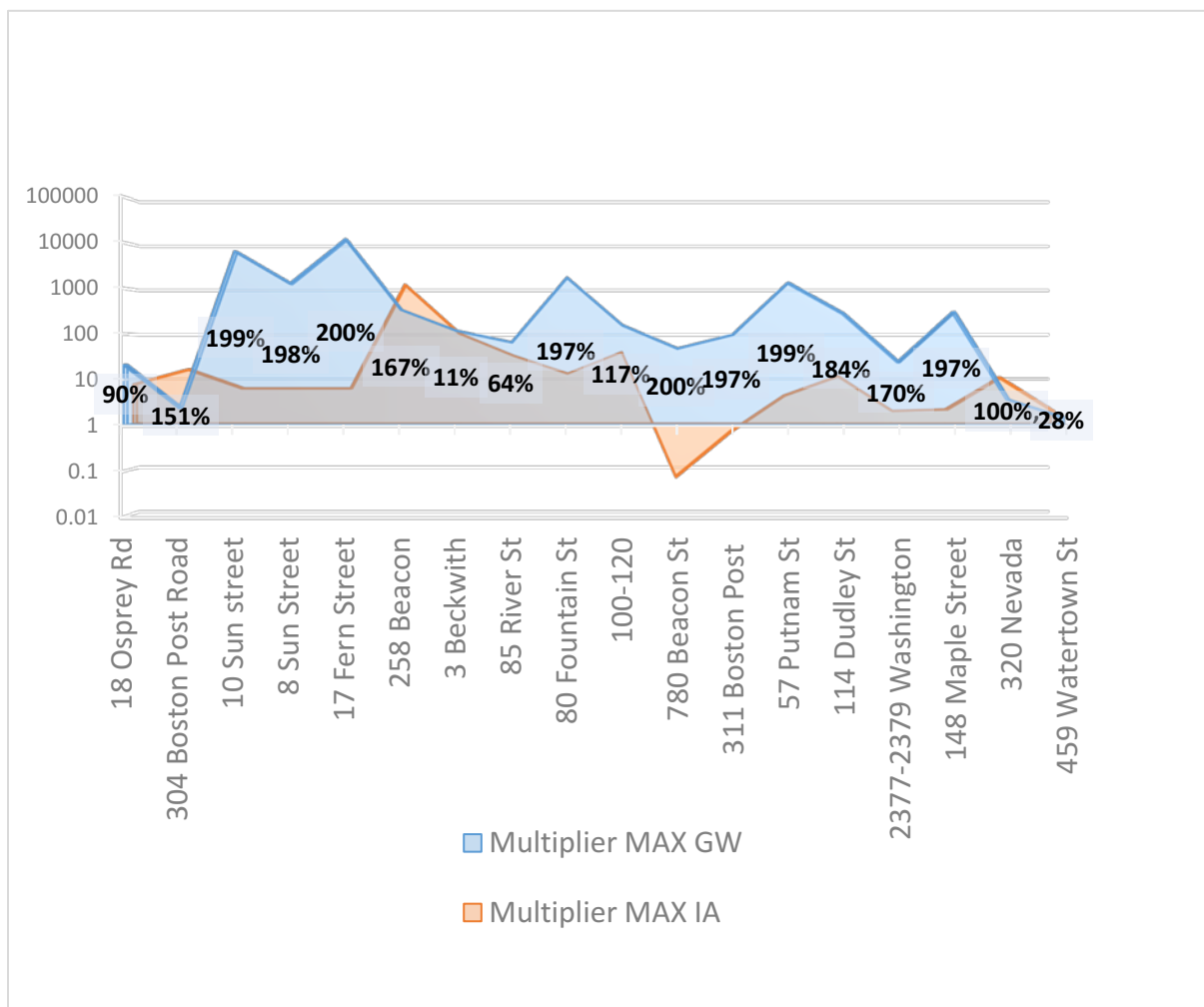


Figure 7. Multiplier of maximum GW & IA sample concentration for each building to applicable regulatory standard.

Results show that reliance on recommended U.S. EPA empirically derived attenuation factors and the attenuation value used by MassDEP in its calculation of the regulatory GW-2 standard for predicting indoor air concentrations of TCE could under-predict vapor intrusion in some circumstances. Approximately 10% of all groundwater to indoor air attenuation factors were higher than recommended both MassDEP and U.S. EPA and over 20% of all subsurface soil gas to indoor air attenuation factors were higher

than recommended both MassDEP and U.S. EPA. Consequently, using groundwater to indoor air attenuation factor demonstrated to be more protective.

Summary and Conclusions

The analyses of these compiled real-world data demonstrated that attenuation factors can be highly variable. The calculated ranges of groundwater to indoor air attenuation values spanned seven orders of magnitude and subsurface soil gas to indoor air attenuation values/factors spanned six orders of magnitude. This range can be attributed to spatial and temporal variability and/or non-representativeness of the subsurface and indoor air samples that were paired for attenuation factor calculations.

This analysis does not explain the factors that contribute to the variability in attenuation factors, but acknowledges the fact that more data are needed to understand variability. Measurements of groundwater and indoor air concentrations should be temporally collected and correlated, but looked at during different seasons to establish reliable long term average attenuation factors, which could provide less conservative and more accurate screening levels for vapor intrusion.

This study examined relationships between subsurface sources and indoor air, where empirically derived attenuation factors from the database were compared with default or recommended attenuation factors. Of all attenuation factors in the database approximately 10% of all groundwater to indoor air attenuation factors were higher than recommended, both by MassDEP and U.S. EPA, and over 20% of all subsurface soil gas to indoor air attenuation factors were higher than recommended by both MassDEP and U.S. EPA, under-predicting indoor air concentrations. Even though investigation of the

vapor intrusion pathway can be a challenge for regulators, the pathway can be adequately controlled or eliminated in a cost-effective manner: sealing cracks, venting sumps, modifying building systems operations and installing SSD system (Fitzgerald, 2009).

Exposure to TCE from vapor intrusion is involuntary and beyond the building occupant's control. It is imperative that investigations be done with the goal of protecting public health. This work demonstrates that attenuation factors used by MassDEP are by and large consistent with this goal, but there were situations where concentrations in indoor air were under-predicted.

Chapter III

Discussion

Overall, this study intended to provide a better understanding of the vapor intrusion phenomenon using real field data with focus on trichloroethylene - an important chemical for vapor intrusion studies because of its extensive use as an industrial degreaser. This has resulted in substantial groundwater and soil contamination not only in the Commonwealth of Massachusetts, but throughout the United States, as evidenced by the number of hazardous waste sites containing TCE (ATSDR, 2014). Additionally, it is still present in some consumer products and therefore can be present in indoor air even in the absence of vapor intrusion (U.S. EPA, 2015-b). A strength of this study is the use of empirically derived data with stringent inclusionary criteria that allows better pairing of samples in the dataset. The research effort adds to the scientific evaluation of current Commonwealth of Massachusetts and federal vapor intrusion regulations and guidance, and despite a sole focus on TCE, the results of this work address broader implications on whether groundwater is an adequate predictor of indoor air exposure.

Both the primary and secondary hypotheses are confirmed, where relationships between subsurface sources and indoor air were examined and empirically derived attenuation factors from the database were compared with recommended attenuation factors. Out of all attenuation factors in the database approximately 10% of all groundwater to indoor air attenuation factors were higher than recommended by both MassDEP and U.S. EPA and over 20% of all subsurface soil gas to indoor air attenuation

factors were higher than recommended by both MassDEP and U.S. EPA, under-predicting indoor air concentrations in some cases. Overall, recommended attenuation factors for TCE are adequate predictors of indoor air concentrations and majority of attenuation factors fell within the range of recommended values. This study concludes that an increase or decrease in groundwater concentration of TCE is not correlated with increases or decreases in indoor air concentration of TCE, indicating that the lack of correlation may be attributed to the site-specific features that are not captured in simple linear regression analysis.

This thesis research demonstrates that attenuation factors can be highly variable and scattered; as shown by calculated ranges of groundwater to indoor air attenuation values span seven orders of magnitude and subsurface soil gas to indoor air attenuation factors span six orders of magnitude. This range can be attributed to spatial and temporal variability and/or non-representativeness of the subsurface and indoor air samples that were paired for attenuation factor calculations. This analysis does not explain the factors that contribute to the variability in attenuation factors, but acknowledges that more data are needed where sampling is done to consider variability i.e. measurements of groundwater and indoor air concentrations should be temporally collected and correlated but looked at during different seasons over time to establish reliable long term average attenuation factors. That in turn would provide less conservative and more accurate screening levels for vapor intrusion.

Most vapor intrusion cases will have different mitigation practices, where costs vary and can range from less than \$1 per square foot (/sq.ft.) for simple ventilation approaches to as much as \$70/sq.ft. for sites with significant access issues and higher

construction labor costs (Kilmer et al., 2016). For new construction, vapor intrusion prevention can be achieved through available VI protection materials and water proofing and both are becoming more available in recent years; in some states, VI mitigation practices are required by the building code. Public health related costs that are being offset by VI mitigation approaches have not yet been quantified but are predicted to be more cost effective compared to long term health consequences of inhaling contaminated vapors.

Research Limitations and Further Study

A major limitation of this study is that all evaluated data were extracted from environmental reports that were produced by others, necessitating the need to accept the information and sample concentrations as accurate and reliable. Additionally, despite the aim to use paired indoor air and groundwater samples that were taken no greater than two years apart, there were a few exceptions where a longer duration between indoor air and groundwater sampling time frame was allowed when necessary data were not available in the environmental reports. Also, this study does not take into account soil types, underground utilities, and building characteristics in data analyses that influence vapor migration from subsurface sources into the indoor air (Pennell 2016; Bekele 2015; Yao 2013; Shen 2013).

During the design of this research, inclusion of soil types as a variable was not possible since not all environmental reports reported soil type data, and where it was reported, all soils were of the same type. Since vapor intrusion is a nationwide problem, more studies are needed that take into account regional and national environmental

characteristics as well as building specifications to better understand the phenomenon. Only a handful of studies have been conducted that rely on empirically collected data relating them to state or federal regulations and guidance. Further research on the relationship between subsurface sources and indoor air with temporally correlated samples over time would help to establish reliable long-term average attenuation factors.

Conclusions

Exposure to TCE from vapor intrusion is involuntary and beyond the building occupant's control. It is imperative that VI investigations be done with the goal of protection of public health. This work demonstrates that attenuation factors used by MassDEP are by and large consistent with this goal but there were situations where concentrations in indoor air were under-predicted. Even though investigation of the vapor intrusion pathway can be a challenge for regulators, the pathway can be adequately controlled or eliminated in a cost-effective manner: sealing cracks, venting sumps, modifying building systems operations and installing SSD system (Fitzgerald, 2009).

In conclusion, this thesis contributes to the VI research field and is of value to state and federal policymakers, as well as professionals that conduct site assessments.

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