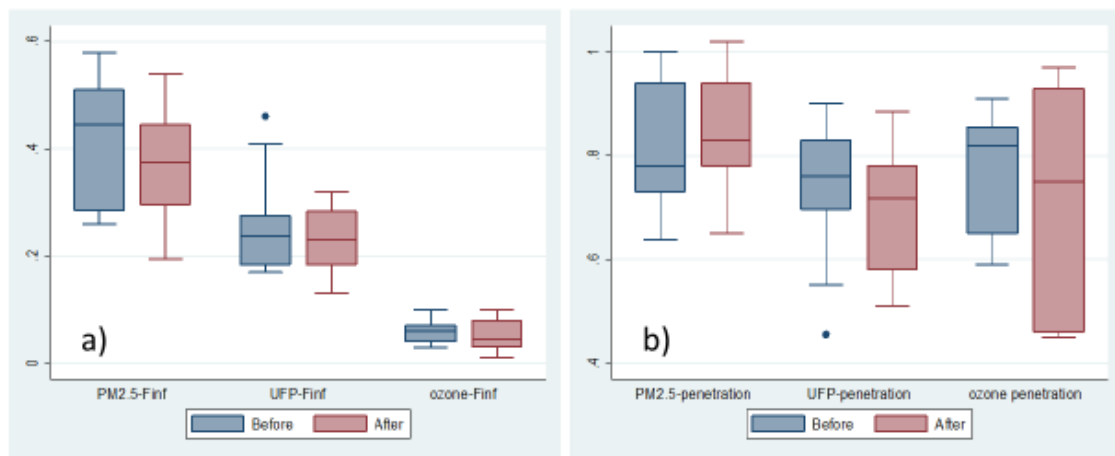


MÁSTER UNIVERSITARIO EN INGENIERÍA INDUSTRIAL

# TRABAJO FIN DE MASTER

## ***OZONE AND FINE AND ULTRAFINE PARTICULATE MATTER PENETRATION INTO RESIDENCES IN CHICAGO***



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## DATOS BÁSICOS DEL PROYECTO

Gran parte de la exposición humana al ozono y a las partículas de origen exterior ocurre en interiores, especialmente en edificios residenciales. El factor de penetración de estos contaminantes a través de las filtraciones en la fachada de un edificio es un parámetro clave que determina su infiltración y permanencia en el interior. Sin embargo, los datos experimentales sobre los factores de penetración de ozono y partículas en edificios reales son limitados. En estudios previos, se ha trabajado para mejorar los métodos de medición de los factores de penetración de ozono, PM2.5 y partículas ultrafinas (UFP) en viviendas bajo condiciones de infiltración.

En este trabajo, se ha diseñado un sistema de instrumentación combinado y se han aplicado estos métodos en una muestra diversa de 22 viviendas en Chicago, IL, con distintas características en la envolvente de los edificios, abarcando tanto casas unifamiliares como multifamiliares. También se han evaluado algunas viviendas antes y después de realizar mejoras en la eficiencia energética. Los factores de penetración promedio ( $\pm$  DE) para PM2.5, UFP y ozono en casas unifamiliares fueron  $0.80 \pm 0.09$ ,  $0.70 \pm 0.11$  y  $0.73 \pm 0.16$ , respectivamente, mientras que en las viviendas multifamiliares los valores fueron de  $0.90 \pm 0.13$ ,  $0.75 \pm 0.16$  y  $0.71 \pm 0.23$ , respectivamente. En 13 de las viviendas, donde se realizaron mediciones tanto antes como después de las mejoras, no se observaron cambios consistentes en los factores de penetración del ozono, PM2.5 o UFP. Sin embargo, las mejoras más comunes, como el aislamiento de áticos y el sellado de aire, solo aumentaron la hermeticidad en un promedio del 14% (con un rango de entre 0% y 46%).

**Palabras clave:** PM2.5, partículas ultrafinas, ozono, factor de penetración, envolvente de edificios, mejoras de eficiencia energética.

## PROIEKTUAREN OINARRIZKO DATUAK

Gizakiak ozonoarekin eta kanpoko partikulekin duen esposizioaren zati handi bat barnealdetan gertatzen da, bereziki bizitegi-eraikinetan. Eraikin baten fatxadako iragazketen bidez kutsatzaile horiek sartzeko faktorea funtsezko parametroa da, haren infiltrazioa eta barruan jarraitzea zehazten duelako. Hala ere, eraikin errealetan ozonoa eta partikulak sartzeko faktoreei buruzko datu esperimentalak mugatuak dira. Aldez aurreko azterketetan, ozonoa, PM2.5 eta partikula ultrafinak (UFP) infiltrazio-baldintzetan dauden etxebizitzetan sartzeko faktoreak neurtzeko metodoak hobetzeko lan egin da.

Lan honetan, instrumentazio konbinatuko sistema bat diseinatu da, eta metodo horiek aplikatu dira Chicagoko (IL) 22 etxebizitzako lagin batean, eraikinen inguratzailean ezaugarri desberdinak dituen, familia bakarreko etxeak zein familia anitzekoak barne hartuta. Halaber, zenbait etxebizitza ebaluatu dira energia-eraginkortasuna hobetu aurretik eta ondoren. Familia bakarreko etxeetan PM2.5, UFP eta ozonorako batez besteko sartze-faktoreak ( $\pm$  DT)  $0.80 \pm 0.09$ ,  $0.70 \pm 0.11$  eta  $0.73 \pm 0.16$  izan ziren, hurrenez hurren; familia anitzeko etxebizitzetan, berriz, balioak  $0.90 \pm 0.13$ ,  $0.75 \pm 0.16$  eta  $0.71 \pm 0.23$  izan ziren, hurrenez hurren.

Etxebizitzetako 13tan, non hobekuntzak egin aurretik zein ondoren neurketak egin baitziren, ez zen ikusi aldaketarik ozonoaren sartze-faktoreetan, PM2.5 edo UFPn. Hala ere, hobekuntza ohikoenek, hala nola atikoen isolamenduak eta airearen zigilatzeak, hermetikotasuna % 14 baino ez zuten handitu batez beste ( % 0 eta % 46 arteko tartearekin).

**Gako-hitzak:** PM2.5, partikula ultrafinak, ozonoa, sartze-faktorea, eraikinen inguratzailea, energia-eraginkortasunaren hobekuntzak.

## BASIC PROJECT DATA

A significant portion of human exposure to outdoor-origin ozone and particulate matter occurs indoors, particularly within residential buildings. The penetration factor of these pollutants through leaks in a building's exterior envelope is a critical factor determining their infiltration and persistence indoors. However, experimental data on the penetration factors of ozone and particles in real-world buildings remain scarce. In previous studies, efforts were made to refine methods for measuring the penetration factors of ozone, PM<sub>2.5</sub> and ultrafine particles (UFPs) in residential settings under infiltration conditions. In this study, a combined instrumentation system was developed and applied to assess 22 homes in Chicago, IL, which exhibited a range of building envelope characteristics. The sample included both single-family and multi-family residences, with a subset evaluated before and after energy efficiency retrofits. Average penetration factors ( $\pm$  SD) for PM<sub>2.5</sub>, UFPs and ozone in single-family homes were  $0.80 \pm 0.09$ ,  $0.70 \pm 0.11$  and  $0.73 \pm 0.16$ , respectively, while in multi-family homes, these values were  $0.90 \pm 0.13$ ,  $0.75 \pm 0.16$  and  $0.71 \pm 0.23$ , respectively. In a subset of 13 homes, pre- and post-retrofit measurements revealed no significant changes in penetration factors for ozone, PM<sub>2.5</sub> or UFPs. However, the retrofits, which predominantly involved attic insulation and air-sealing, resulted in an average increase in airtightness of only 14% (ranging from 0% to 46%)

**Keywords:** PM<sub>2.5</sub>, ultrafine particles, ozone, penetration factor, building envelope, energy efficiency retrofit.

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# 1.- INTRODUCTION

Human exposure to ambient pollutants such as particulate matter, ozone and nitrogen oxides has been consistently linked to various adverse health outcomes in epidemiological studies. However, a significant portion of this exposure occurs indoors, particularly within residential buildings where individuals spend the majority of their time. This indoor exposure is critically influenced by the building's ability to filter outdoor air as it enters the indoor environment. Despite its importance, there has been a scarcity of measurements regarding the penetration of outdoor pollutants into real indoor settings, primarily due to the high costs, uncertainties and disruptions to building occupants associated with current methods.

Epidemiological studies have repeatedly demonstrated the harmful effects of elevated outdoor pollutant levels on human health. For instance, higher concentrations of fine particulate matter (PM<sub>2.5</sub>, particles smaller than 2.5 µm in diameter) have been correlated with increased risks of respiratory issues, mortality and lung cancer ((Brook et al., 2010); (Dockery et al., 1993); (E. Chen & Miller, 2007); (Pope et al., 2002); (Pope et al., 2009); (Pope & Dockery, 2006)). Similarly, elevated levels of ultrafine particles (UFPs, particles smaller than 100nm diameter) have been associated with higher rates of cardiorespiratory diseases and mortality ((Osunsanya et al., 2001); (Penttinen et al., 2001); (Stölzel et al., 2006); (von Klot et al., 2002); (Weichenthal et al., 2007)). Additionally, increased ambient ozone levels have been linked to a range of negative health outcomes, including hospital admissions, respiratory illnesses and short-term mortality ((Bell et al., 2004); (Dominici et al., 2006); (Bell et al., 2014); (Fann et al., 2012); (Jerrett et al., 2009); (Henschke et al., 2012); (Hubbell et al., 2005); (Ito et al., 2005); (Gent et al., 2003)). Ozone also plays a crucial role in indoor chemistry (Fadeyi et al., 2013); (Shu & Morrison, 2012); (Wang & Waring, 2014); (Waring & Siegel, 2013); (Waring & Wells, 2015)), suggesting that exposure to the byproducts of ozone reactions could contribute to various adverse health effects ((WESCHLER\* & SHIELDS, 2000), (Weschler, 2006a)). Furthermore, elevated concentrations of nitrogen oxides (NO<sub>x</sub>),

including oxide (NO) and nitrogen dioxide (NO<sub>2</sub>), have been associated with a broad spectrum of health issues, such as respiratory and cardiovascular problems, lung cancer and increased mortality ((Shi et al., 2016)).

While outdoor pollutant concentrations are often used as proxies for human exposure in epidemiological studies, it is essential to understand indoor exposures to outdoor particles and gas-phase pollutants like ozone and nitrogen oxides. This is particularly important, because:

1. People spend most of their time indoors, with approximately 70% of that time spent at home ((Klepeis et al., 2001)).
2. Outdoor particles, ozone and NO<sub>x</sub> can infiltrate and persist within indoor environments ((Fabian et al., 2012); (C. Chen & Zhao, 2011); (Zota et al., 2005); (Dimitroulopoulou et al., 2001); (Weschler, 2006a); (Avol et al., 1998a)).

As a result, the majority of human exposure to outdoor particles, ozone and nitrogen oxides actually occurs indoors, particularly in residential settings where individuals spend most of their time ((Z. Yang et al., 2015); (C. Chen et al., 2011a); (Baxter et al., 2007); (Baxter et al., 2006a); (Bhangar et al., n.d.); (Weschler et al., 2006); (Qing et al., 2004a)). Therefore, a deeper understanding of the transport mechanisms of outdoor particles, ozone and NO<sub>x</sub> into indoor residential environments is crucial for enhancing our comprehension of human exposure to these pollutants.

## 2.- OBJECTIVES

The primary aim of this dissertation is to address the significant gap in our understanding of how outdoor pollutants infiltrate indoor environments, specifically within residential buildings. This research is guided by the overarching objective to develop and apply novel methods to characterize the transport of outdoor particles, ozone and nitrogen oxides (NO<sub>x</sub>) into residential buildings. Current studies on pollutant infiltration are limited due to challenges in experimental methodologies, high instrumentation costs and the invasive nature of measurements, which often disrupt the lives of building occupants. Additionally, existing studies frequently overlook key building details that could be critical for predicting infiltration results.

To overcome these limitations, the research is structured around two central objectives:

1. Development and refinement of measurement methods: this objective focuses on innovating and refining methodologies for accurately measuring the penetration of outdoor pollutants – specifically particulate matter, ozone and nitrogen oxides – into indoor environments. The goal is to create cost-effective, minimally invasive and accurate tools capable of operating under various building conditions. By addressing the limitations of current techniques, this research aims to enhance our ability to quantify pollutant infiltration in a way that is both practical and precise.
2. Application to a diverse sample of residential units: the second objective is to apply these refined measurement methods to a representative sample of residential buildings in Chicago, IL. This application will include a variety of building types, such as single-family homes, multifamily homes and homes both before and after undergoing energy efficiency retrofits. By selecting a diverse range of buildings, this research aims to capture a comprehensive understanding of how different building characteristics influence the infiltration of outdoor pollutants. The findings will provide critical data that can be used to improve models assessing indoor exposure to outdoor

pollutants, thereby contributing valuable insights to the field of environmental health.

These objectives are designed to advance our ability to quantify and mitigate human exposure to harmful outdoor pollutants within residential environments. The outcomes of this research will not only enhance exposure assessment techniques but also support the development of better-informed public health policies and building design practices aimed at protecting indoor air quality. The overarching research objectives are divided into four sub-objectives:

## 2.1.- Refine methods to accurately measure envelope penetration factors for ozone

In the only known study examining ozone penetration, a calibrated fan was used to depressurize houses, artificially increasing the AER and raising the steady-state indoor ozone concentration above the detection limit of the UV photometric ozone monitor available at the time. This monitor, which relied on UV absorption at 254 nm, was prone to interference from mercury, water vapor and various indoor VOCs, including styrene, methylstyrene, o-cresol, nitrocresol and other aromatic compounds with electron-withdrawing groups (e.g., -OH, -NO<sub>2</sub> and -CHO) ((Johnson et al., 2014); (Ollison, Crow, et al., 2013); (Spicer et al., 2010); (H. Zhao & Stephens, 2016a); (Parrish et al., 2009); (Wilson & Birks, 2006); (Grosjean & Harrison, 1985a); (Huntzicker & Johnson, 1979)). These interferences may have affected indoor detection limits, leading to the use of the blower door method to artificially elevate indoor concentrations. Additionally, the reliance on artificially high AERs in this method may have produced results that do not accurately reflect real-world conditions due to changes in airflow patterns through leakage pathways.

To address these limitations, this research refines an existing method for measuring ozone penetration factors in residences under natural infiltration conditions, without the use of artificial depressurization. A new NO-scrubbed ozone monitor, with a lower detection limit and reduced susceptibility to interference, is utilized. The method is applied in an unoccupied, sparsely furnished test apartment on the

campus of the Illinois Institute of Technology in Chicago, IL. Repeated measurements of ozone penetration factors are conducted, leading to analyses that include:

1. An assessment of the accuracy and repeatability of the test method using various mathematical approaches to determine ozone decay rate constants and penetration factors.
2. Side-by-side comparisons between two ozone monitors during natural infiltration tests, specifically comparing an “interference-free” monitor with a conventional UV absorbance ozone monitor previously used.
3. Comparisons of results from natural infiltration experiments with those conducted using a blower door, to validate the originally published method.

## 2.2.- Refine methods to more rapidly and accurately measure envelope penetration factors for fine and ultrafine particles

Field experiments on ambient particle penetration have been conducted by only a limited number of researchers and are rarely performed due to their lack of standardization, complexity and high costs. Some studies have relied on overnight measurements to estimate penetration factors during periods when occupants were presumed to be inactive ((Long et al., 2001); (Vette et al., 2001)). Others have extended measurement durations to two or more days while the building remained unoccupied (Rim et al., 2010a). Notably, no studies have experimentally characterized PM<sub>2.5</sub> penetration factors, although several have estimated the penetration factor (P) for PM<sub>2.5</sub> using time-integrated gravimetric measurements and various statistical techniques ((Qing et al., 2004b), (Q. Y. Meng et al., 2009); (Riediker et al., 2003a)).

In response to these challenges, objective 2 of this research focuses on developing a method for rapidly measuring size-resolved particle penetration factors for fine and ultrafine particles in residential settings using portable particle sizing instrumentation. Replicate measurements are conducted in an unoccupied apartment unit to:



1. Evaluate the accuracy and repeatability of the method in determining both penetration factors and deposition loss rate constants for size-resolved particles, as well as integral measures of UFPs and PM<sub>2.5</sub>.
2. Perform side-by-side comparisons of PM<sub>2.5</sub> infiltration factors measured using different aerosol instruments.
3. Investigate potential influences of indoor and outdoor environmental factors on both penetration factors and deposition loss rate constants.

### 2.3.- Develop novel methods to measure envelop penetration factors for nitrogen oxides

Nitrogen oxides (NO<sub>x</sub>) are criteria pollutants that can react with various building enclosure materials ((Grøntoft & Raychaudhuri, 2004); (Spicer et al., 1989)). However, the penetration efficiency of NO<sub>x</sub> through building envelopes has not been thoroughly investigated due to several challenges:

1. Many studies on indoor and outdoor NO<sub>x</sub> rely on passive integrated samplers, which do not provide the time-resolved data needed to estimate first-order decay rates for NO<sub>x</sub>.
2. Studies that have measured time-resolved indoor NO<sub>x</sub> often cannot differentiate between indoor and outdoor sources.
3. Most time-resolved NO<sub>x</sub> measurement instruments use a chemiluminescence reaction method with high detection limits, which is susceptible to interference from indoor species such as HONO, HNO<sub>3</sub> and peroxyacyl nitrates ((Kebabian et al., 2008); (McClenny et al., 2002)), making accurate time-resolved indoor/outdoor measurements difficult for estimating penetration factors (P) and decay rates (k).

In response, objective 3 of this research focuses on developing new methods to measure envelope penetration factors for NO<sub>2</sub>/NO/NO<sub>x</sub> using a UV absorption monitor that offers lower detection limits and reduced susceptibility to interference compared to commonly used NO<sub>x</sub> instrumentation. These methods are developed and tested through experiments conducted in an unoccupied and sparsely



furnished test apartment on the campus of Illinois Institute of Technology in Chicago, IL. Repeated measurements of NO<sub>2</sub>/NO/NO<sub>x</sub> penetration factors are collected, providing a dataset for the following analyses:

1. Evaluation of the accuracy and repeatability of the test method in determining decay rate constants and penetration factors.
2. Investigation of potential influences of indoor and outdoor environmental factors on both penetration factors and deposition loss rate constants.

## 2.4.- Conduct field measurements of outdoor pollutant penetration in a diverse sample of residential buildings in Chicago, IL

Previous modeling and experimental studies have indicated that pollutant penetration may be influenced by various building envelope characteristics, such as crack geometry, indoor-outdoor pressure differences, building façade materials and air leakage coefficients ((Stephens & Siegel, 2012a); (Liu & Nazaroff, 2003a); (Saliy Liu et al., 1995)).

In objective 4, the developed and refined test methods are applied to a diverse sample of 23 existing homes in Chicago, IL. This sample includes single-family residences and multi-family units with a variety of construction ages and envelope material types. Additionally, given the increasing application of energy efficiency retrofits, such as air-sealing and insulation, which are intended to reduce energy consumption but may alter air infiltration pathways ((Nabinger & Persily, 2011); (McCold et al., 2008); (Schweitzer & Eisenberg, n.d.)), a subset of the field measurements will involve testing homes both before and after undergoing energy retrofits. These measurements aim to enhance understanding of the variability in factors that govern indoor concentrations of ambient pollutants in homes with differing building characteristics.



## 3.- CONTEXT

The transport of outdoor pollutants into indoor environments is a complex process influenced by various factors. The infiltration factor ( $F_{inf}$ ) refers to the fraction of outdoor pollutants that penetrate and remain indoors in the absence of indoor source. This process is critical for understanding human exposure to pollutants such as particulate matter, ozone and nitrogen oxides (NO<sub>x</sub>), especially in residential settings where people spend most of their time. Studying the factors that influence this process, such as air exchange rates, deposition loss rates and penetration factors, is essential to improving our understanding of indoor air quality and its health impacts. This chapter reviews previous studies and methodologies for measuring these parameters, emphasizing the need for continued development of precise techniques to assess the transport of outdoor pollutants into homes more effectively.

### 3.1.- Outdoor pollutants transport indoors

The infiltration factor ( $F_{inf}$ ) represents the portion of outdoor pollutants that infiltrate into indoor environments. It is defined as the ratio of indoor concentration in the absence of any indoor sources, under steady-state or time-averaged conditions. The following image illustrates a simplified schematic of how ambient pollutants, including particulate matter, ozone and nitrogen oxides (NO<sub>x</sub>) are transported into a typical residential building without indoor sources.

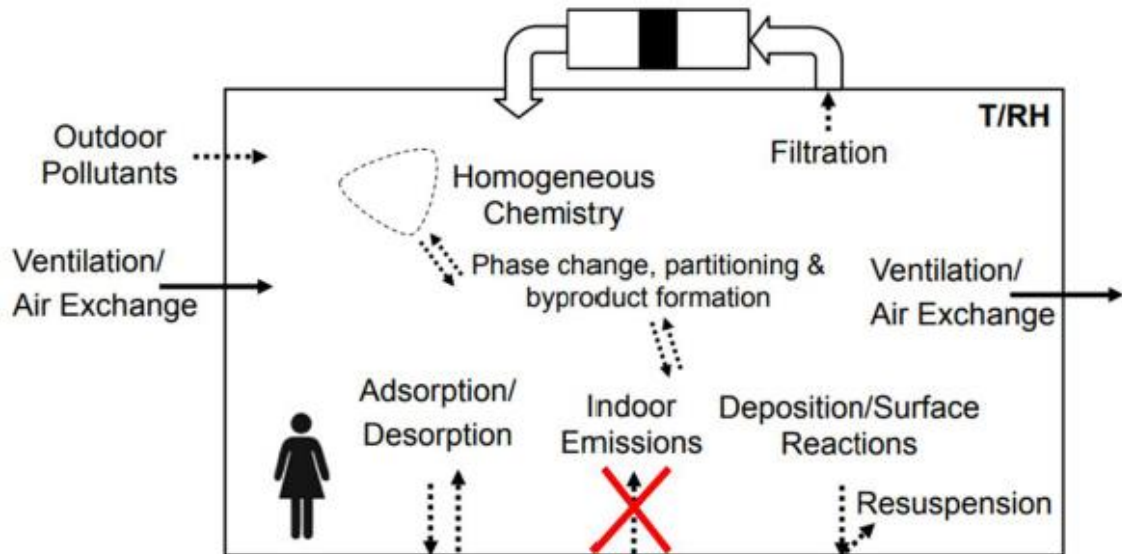


Figure 1.- Simplified schematic showing the transport of ambient pollutants into a typical residential building in the absence of indoor sources.

Three key parameters determine the infiltration factor:

1. The air exchange rate ( $\lambda$  or AER,  $\text{h}^{-1}$ )
2. The particle deposition loss rate constant ( $k$ ,  $\text{h}^{-1}$ ). This parameter accounts for the removal of pollutants due to deposition on interior surfaces
3. The penetration factor ( $P$ , dimensionless), which is the fraction of pollutants in the infiltrating air that successfully passes through the building's enclosure.

The time-averaged factor for a specific pollutant in a building is expressed in Equation 1:

$$\frac{C_{in}}{C_{out}} \Big|_{\text{no indoor source}} = F_{inf} = \frac{P\lambda}{\lambda + k}$$

Equation 1.

### 3.2.- Infiltration factor

The infiltration factor ( $F_{inf}$ ) represents the equilibrium fraction of ambient pollutants that penetrate indoors and remain airborne. Measuring  $F_{inf}$  is relatively straightforward when indoor sources are minimal or absent. Such measurements

have been conducted across a wide range of homes under various operational conditions for different classes of particulate matter (e.g., (Ramos et al., 2015); (L. A. Wallace et al., 2010); (Macneill et al., 2014); (R. W. Allen et al., 2012a); (MacNeill et al., 2012a); (C. Chen et al., 2011b); (Rim et al., 2010b); (Bennett & Koutrakis, 2006); (L. Wallace & Williams, 2005a); (Zhu et al., 2005); (Long & Sarnat, 2004)) as well as ozone (e.g., (Avol et al., 1998b); (Brauner et al., 2014); (Cattaneo et al., 2011a); (K. Lee et al., 1999); (Saliy Liu et al., 1995); (Romieu et al., 1998); (Zhang & Liou, 1994)). However, accurate measurements of  $F_{inf}$  for NO<sub>x</sub> constituents are less common (e.g., (Meier et al., 2015); (Rivas et al., 2015a); (Wichmann et al., 2010); (Baxter et al., 2006b); (W. Yang et al., 2004a); (Kulkarni & Patil, 2002)), partly because indoor sources like cooking and space heating using gas and other fuels complicate the exclusion of indoor influences, making direct  $F_{inf}$  estimation challenging (e.g., (Logue et al., 2014); (Kornartit et al., 2010); (W. Yang et al., 2004b)). Most studies have focused on simultaneous indoor-outdoor measurements of nitrogen oxides (typically NO<sub>2</sub>), generally suggesting that ambient NO<sub>x</sub> infiltration is influenced by factors such as building and household characteristics, human activities, ventilation type and both indoor and outdoor environmental conditions (e.g., (Q. Meng et al., 2012); (Q. Y. Meng et al., 2012); (Physick et al., 2011); (Ballester et al., 2010); (Sørensen et al., 2005); (Zota et al., 2005); (Lai et al., 2004a); (W. Yang et al., 2004b); (K. Lee et al., 2000)). However, specific studies on outdoor NO<sub>x</sub> infiltration factors remain limited.

### 3.3.- Air exchange rate and indoor loss rate

Measuring the air exchange rate (AER) is more complex but still relatively straightforward using standardized tracer gas decay techniques. Air exchange rates have been recorded in thousands of homes ((Murray & Burmaster, 1995)). Similarly, measurements of the deposition loss rate constant (k) are also relatively easy to obtain using an elevation and decay procedure alongside simultaneous AER measurements. Numerous studies have reported particle deposition loss rate constants across various particle sizes and building operational conditions, with and without central HVAC filtration (e.g., (W. C. Lee et al., 2014); (Rim, Wallace, &

Persily, 2013); (Rim et al., 2010b); (B. Zhao & Wu, 2007); (He et al., 2005); (L. A. Wallace et al., 2004); (Thatcher et al., 2002); (Thatcher et al., 2002); (Thatcher & Layton, 1995a)). Similarly, first-order decay rates have been measured in laboratory chambers and various real buildings for both ozone ((Weschler, 2000); (K. Lee et al., 1999)) and NO<sub>x</sub> constituents ((Grøntoft & Raychaudhuri, 2004); (W. Yang et al., 2004b); (Weschler et al., 1994); (Spicer et al., 1989); (Spicer et al., 1993); (Nazaroff & Cass, 1986)).

### 3.4.- Penetration factor

In addition to measuring infiltration factors, air exchange rates and loss rates, accurately determining pollutant penetration factors through building enclosures is critical, especially in U.S. homes. This is particularly important because most residential buildings in the U.S. lack dedicated mechanical ventilation systems, relying instead on a combination of infiltration (i.e., air entering through leaks in the building envelope) and window opening for ventilation (Chan et al., 2005). However, window opening frequencies in U.S. residences are not well documented and typically occur less than 20 – 30% of the time, depending on outdoor weather conditions and occupant preferences ((C. Chen et al., 2012); (El Orch et al., 2014); (Johnson & Long, 2004); (Price & Sherman, n.d.)). Consequently, during much of the year and in many locations, particles, ozone and NO<sub>x</sub> from outdoor sources enter indoor environments primarily through leaks in the building envelope assembly. If the variability in envelope penetration factors is not accounted for, it can lead to inaccurate infiltration factor estimates, ultimately resulting in exposure errors in epidemiological studies ((Breen et al., 2015); (Waring, 2014); (Baxter et al., 2009); (Baxter et al., 2011); (Hodas et al., 2016), (Baxter et al., 2013); (C. Chen et al., 2011c), (C. Chen et al., 2012); (Weschler, 2000), (Weschler, 2006b); (Georgopoulos et al., 2005)). However, measuring envelope penetration factors is generally the most challenging and time-consuming task, as both the penetration factor (P) and the loss rate constant (k) must be estimated from a single mass balance equation during periods without indoor sources but with significant concentration variations.

Due to these challenges, pollutant penetration factor measurements have been conducted in very few buildings worldwide.

Modeling studies have shown that pollutant penetration through building leaks is influenced by several factors, including the geometry of the leaks, pressure differences across the envelope, air velocities through the leaks, building envelope materials (for ozone) and particle size (for particulate matter) ((Liu & Nazaroff, 2001a)). Although previous studies assumed that NO<sub>2</sub>/NO/NO<sub>x</sub> penetrates building enclosures with 100% efficiency ((Physick et al., 2011); (W. Yang et al., 2004b)), laboratory studies have indicated that NO<sub>x</sub> can react with various building materials commonly used in enclosures, suggesting that some NO<sub>x</sub> removal might occur as outdoor air infiltrates indoors ((Grøntoft & Raychaudhuri, 2004); (Spicer et al., 1989)).

To date, no studies have specifically investigated NO<sub>2</sub>/NO/NO<sub>x</sub> penetration factors through building envelopes. Specific measurements of particle penetration factors have been conducted in fewer than 50 homes worldwide ((Rim et al., 2010b), (Rim, Wallace, Persily, et al., 2013); (Stephens et al., 2012); (Zhu et al., 2005); (Chao et al., 2003); (Thatcher et al., 2003); (Long et al., 2001); (Mosley et al., 2001); (Vette et al., 2001); (Thatcher & Layton, 1995a)), and there has been only one study that measured ozone penetration factors in 8 residential buildings in Texas (Stephens et al., 2012). These studies have varied significantly in their measurement approaches, test durations and the resulting uncertainty in parameter estimates. Therefore, this work will focus on refining and developing methods, where applicable, to more rapidly and accurately measure envelope penetration factors for size-resolved particulate matter, ozone and NO<sub>x</sub> in residential buildings.

## 4.- LITERATURE REVIEW

### 4.1.- Particulate matter infiltration factors

Field measurements of particle infiltration factors are relatively straightforward to conduct and have been applied in thousands of homes worldwide, including for ultrafine particles (UFPs) and PM<sub>2.5</sub>. These studies have demonstrated that time-averaged  $F_{inf}$  values can vary widely between residential buildings, ranging from less than 0.1 to nearly 1.0. The variation depends on factors such as particle size/class (with  $F_{inf}$  for UFPs typically being lower than for PM<sub>2.5</sub>), window-opening behavior and other underlying characteristics like envelope airtightness.

The most commonly used method to measure  $F_{inf}$  involves comparing indoor and outdoor particle concentration in the absence of indoor sources. This can be achieved through several approaches:

1. Testing unoccupied houses where no indoor sources are present ((Rim et al., 2010b); (H. S. Lee et al., 1997)).
2. Measuring time-integrated gravimetric indoor/outdoor (I/O) concentrations in occupied houses and applying statistical methods (e.g., linear regression of indoor concentration against outdoor concentration to estimate the slope) to account for indoor sources ((MacNeill et al., 2012b); (Hoek et al., 2008); (Qing et al., 2007); (Sarnat et al., 2006); (Weisel et al., 2005); (L. Wallace & Williams, 2005b); (Lai et al., 2004b); (R. Allen et al., 2003); (L. A. Wallace et al., 2003); (Riediker et al., 2003b); (Lachenmyer & Hidy, 2000); (Gordian et al., 1996); (Barry Ryan et al., 1986); (Dockery & Spengler, 1981)).
3. Using time-resolved I/O concentration ratios during periods with no indoor sources (e.g., nighttime) ((Long et al., 2001)).
4. Measuring the elemental composition of ambient tracers that have no significant indoor sources, such as the I/O sulfur ratio as a surrogate for  $F_{inf}$  for ambient PM<sub>2.5</sub> ((R. W. Allen et al., 2012b); (L. Wallace & Williams, 2005b)).

## 4.2.- Particle penetration factors

The envelope penetration factor is fundamental to any assessment of infiltration factors, as it directly characterizes how pollutants penetrate through the building envelope. Different particle sizes interact with cracks in the building envelope in distinct ways:

- Ultrafine particles ( $< 0.1 \mu\text{m}$ ) tend to deposit on crack walls due to Brownian motion.
- Coarse particles ( $> 1 \mu\text{m}$ ) typically impact or settle by gravitation.
- Mid-sized particles ( $0.1 - 1 \mu\text{m}$ ) are not predominantly influenced by either force ((Liu & Nazaroff, 2001b)).

Specific measurements of particle penetration factors have been conducted in fewer than 50 homes globally. No studies have directly measured integral PM<sub>2.5</sub> penetration factors; however, some research has estimated average penetration factors for building samples using time-integrated gravimetric data and various statistical methods ((Qing et al., 2004c); (R. Williams et al., 2003)). The following sections review previous investigations into the penetration of outdoor particulate matter, primarily conducted through modeling efforts, laboratory experiments and specific field studies.

### 4.2.1.- Modeling studies

Liu & Nazaroff, 2001b modeled the size-resolved penetration of particles through building envelope structures. Their predictions indicated that particles within the  $0.1 - 1 \mu\text{m}$  range would exhibit the highest penetration efficiency through idealized smooth rectangular cracks in a structure. In contrast, the building envelope would filter many super-micron ( $> 1 \mu\text{m}$ ) and ultrafine ( $< 100 \text{nm}$ ) particles through gravitational settling and Brownian diffusion, respectively. The penetration efficiency is theoretically influenced by pressure differences across the crack and the geometry of the crack. For wall cavities, fiberglass insulation was predicted to act as a perfectly efficient particle filter for all particle sizes, regardless of pressure differences.



#### 4.2.2.- Laboratory measurements

Thornburg et al., 2001 conducted particle penetration experiments using a two-compartment chamber separated by a partition with idealized horizontal slits simulating leakage paths. Particles ranging from 0.05 to 5  $\mu\text{m}$  were generated in one compartment and transported to the other via airflow induced by an applied pressure differential. At a pressure of 2 Pa, only 2% of 2  $\mu\text{m}$  particles and less than 1% of 5  $\mu\text{m}$  particles penetrated the envelope structure. Additionally, Liu & Nazaroff, 2003b measured particle concentrations upstream and downstream of various common building materials, including aluminum, brick, concrete, plywood, lumber and oriented strand board, each with different crack geometries. Their findings indicated that particle size and crack height were the primary factors governing particle penetration. The penetration factor was nearly 1 for 0.1 – 1.0  $\mu\text{m}$  particles in cracks over 0.25 mm in height and pressure differences exceeding 4 Pa. However, the practical relevance of these idealized laboratory penetration tests is limited, as particle penetration is likely to be reduced in non-ideal cracks found in real buildings, which typically exhibit significant surface roughness and irregular geometry.

#### 4.2.3.- Field measurements

Field measurements of penetration factors in real residential buildings are challenging and typically require the house to be unoccupied or for indoor sources to be negligible. These measurements also necessitate the use of real-time instruments to simultaneously monitor indoor and outdoor particle number or mass concentrations over time, particularly when the indoor air is reasonably well-mixed. The dynamic mass balance approach models the time-varying indoor particle concentration for the measured size categories, as represented in Equation 2:

$$\frac{dC_{in}}{dt} = P\lambda C_{out} - (\lambda + k)C_{in}$$

Equation 2.

To solve this equation, the air exchange rate (AER) must be estimated concurrently during the test period, which can be done through periodic injections of a tracer gas and measuring its subsequent decay. The AER can be determined using linear regression of the natural logarithm of indoor tracer gas concentrations over time. However, solving Equation 2 remains difficult even with a known AER because there are still two unknown parameters in one mass balance equation. Therefore, three mathematical methods have been commonly used in previous studies to achieve relatively accurate estimates for both  $P$  and  $k$ .

### *Steady-state solution*

When indoor concentrations remain relatively steady compared to outdoor concentrations, the penetration factor can be determined using Equation 3, which is derived from Equation 1, provided that the deposition loss rate is known:

$$P = F_{inf} \frac{\lambda + k}{\lambda}$$

*Equation 3.*

Achieving an accurate estimate of the infiltration factor ( $F_{inf}$ ) typically requires long-term measurements, with particle deposition loss rates measured in advance using an injection and decay method. For instance, Thatcher & Layton, 1995b measured size-resolved particle concentrations (ranging from 0.3 to 25  $\mu\text{m}$  in diameter) indoors and outdoors at a two-story residence, while also measuring the air exchange rate (AER) with tracer gas decay. They determined size-resolved deposition rates by artificially elevating indoor aerosol concentrations and solving for the subsequent decay rate, after accounting for the AER. Size-resolved penetration factors were then estimated using steady-state indoor/outdoor concentration ratios, measured AER and the previously estimated decay rate in conjunction with Equation 2. For this residence, the penetration factors were approximately 1 for all particle sizes reported.

In a similar test conducted by Vette et al., 2001 in a single-family residence using overnight measurements, the size-resolved penetration factors ranged from

approximately 0.5 to 0.8 for particles between 0.001 and 2.5  $\mu\text{m}$ . Long et al., 2001 further refined this method by using a linear regression form of Equation 2, with the inverse of the air exchange rate as a dependent variable. This study reported that the penetration efficiency of particles ranging from 0.02 to 10  $\mu\text{m}$  in 9 homes varied between approximately 0.2 and greater than 0.9, depending on particle size, season and home characteristics.

### *Analytical solution*

When outdoor particle concentrations remain relatively constant, Equation 2 can be simplified to an analytical solution using a constant average outdoor concentration, as shown in Equation 4:

$$C_{in,t} = C_{in,t=0}e^{-(\lambda+k)t} + \frac{P\lambda\overline{C_{out}}}{\lambda + k}(1 - e^{-(\lambda+k)t})$$

*Equation 4.*

Estimates of P and k can be obtained through non-linear least squares regression of the data against the analytical solution. This method generally provides estimates of P and k with relatively low standard error but may be limited when outdoor concentrations fluctuate during the test period (H. Zhao & Stephens, 2016b). Consequently, it is typically applied to shorter-term measurements.

For instance, Chao et al., 2003 conducted short-term tests in six non-smoking, unoccupied apartments. Indoor particle concentrations in the test apartments were first elevated through natural ventilation, then the windows and doors were closed to measure the subsequent decay of indoor particles under infiltration conditions. Outdoor particle concentrations were recorded before and after the test. Size-resolved penetration factors (ranging from 0.02 to 1  $\mu\text{m}$ ) were calculated by applying Equation 4 to the natural decay curves of indoor particle concentrations. The average penetration factors ranged from approximately 0.6 (standard deviation  $\sim$  0.3) for particles between 0.01 and 1.0  $\mu\text{m}$ , to approximately 0.7 – 0.8 (standard deviation  $\sim$  0.2) for particles between 0.5 and 2.5  $\mu\text{m}$ , and approximately 0.5 (standard deviation  $\sim$  0.3) for particles between 2.6 and 10  $\mu\text{m}$ .

### *Discretized solution or forward-marching solution with time step*

Because outdoor concentrations may fluctuate during the test periods, Equation 2 can be adapted into a discretized solution form, allowing it to be fitted to data that reflect indoor responses to changes in outdoor concentrations at each measured time step, as shown in Equation 5:

$$C_{in,t} = P\lambda C_{out,t}\Delta t + (1 - (\lambda + k)\Delta t)C_{in,t-1}$$

*Equation 5.*

Estimates of P and k can be derived using non-linear least squares regression between Equation 5 and the measured data. This approach accommodates varying outdoor particle concentrations during the test period, generally resulting in lower uncertainties when outdoor levels fluctuate. However, the time step  $\Delta t$  should be sufficiently small relative to the total test period to ensure accurate estimates based on discretized data points.

For instance, Thatcher et al., 2003 conducted size-resolved particle measurements (0.1 – 1  $\mu\text{m}$ ) indoors and outdoors at two houses while simultaneously measuring air exchange rates (AER) using tracer gas decay. Their experimental approach involved:

1. Measuring the decay rate of particles after artificially elevating concentrations (achieved by igniting a natural gas burner and performing vigorous re-suspension activities).
2. Rapidly reducing particle concentrations below background levels by introducing HEPA-filtered outdoor air.
3. Measuring the subsequent particle concentration rebound period and applying it to Equation 5 to determine both penetration factors and deposition loss rates. Estimated penetration factors ranged from approximately 1 for 0.1  $\mu\text{m}$  particles to around 0.3 for 10  $\mu\text{m}$  particles.

Zhu et al., 2005 measured size-resolved ultrafine particle concentrations in four apartment units, without cooking or cleaning activities, over six consecutive days.

They used an SMPS combined with a switch manifold to measure alternating indoor/outdoor concentrations at 9-minute intervals. Applying Equation 5 with a 20-minute time step, they reported relatively constant penetration factors of approximately 0.5 for 0.02 – 0.2  $\mu\text{m}$  particles, with average penetration factors declining for smaller particles, down to less than 0.2 for particles smaller than 0.01  $\mu\text{m}$ .

Similarly, Rim et al., 2010b measured alternating indoor/outdoor ultrafine particle (< 0.1  $\mu\text{m}$ ) concentrations at 2.5-minute intervals for 60 hours in an unoccupied test house under two conditions:

1. With closed windows
2. With a window open approximately 7.5 cm.

The penetration factor increased from approximately 0.2 for 0.01  $\mu\text{m}$  particles to an asymptote of about 0.6 for 0.03 – 0.1  $\mu\text{m}$  particles with closed windows, and ranged from 0.6 to 0.8 across all particle sizes with the window open.

Stephens & Siegel, 2012b developed a refined method for measuring submicron particle penetration factors in 19 single-family houses using the elevation and decay strategy from C. Chen et al., 2011d. Particle deposition loss rates were initially estimated using log-linear regression applied to the early portion of indoor natural decay, and these estimates were the used in Equation 5 to determine penetration factors with relatively high accuracy. The study reported a mean ( $\pm$ standard deviation) penetration factor for submicron particles (20 – 100 nm, not size-resolved) of  $0.47 \pm 0.15$  in residences relying on infiltration for ventilation, ranging from  $0.17 \pm 0.03$  to  $0.72 \pm 0.08$ .

### 4.3.- Ozone indoor/outdoor ratio and infiltration factor

Indoor concentrations of ozone originating from outdoors are influenced by the air exchange rate (AER), penetration through the building envelope, and the first-order ozone decay rate, which accounts for the overall indoor loss due to heterogeneous and/or homogeneous reactions. Given that in most buildings, indoor ozone is

primarily transported from outdoors (Weschler, 2006c), the long-term indoor-outdoor (I/O) ozone concentration ratio can serve as a reliable measure of the infiltration factor.

Weschler, 2000 compiled the majority of known measured I/O ozone concentration ratios from various types of buildings reported in the literature. These ratios typically range from 0.05 in tightly sealed buildings or those utilizing activated carbon HVAC filtration, to 0.85 in mechanically ventilated buildings with very high air exchange rates. Focusing on large residential studies, Avol et al., 1998b and Romieu et al., 2012 reported average I/O ratios of  $0.37 \pm 0.25$  and  $0.20 \pm 0.18$  in 126 U.S. homes and 145 homes in Mexico, respectively. In a smaller study, Zhang & Liroy, 1994 found average I/O ratios ranging from  $0.22 \pm 0.09$  to  $0.62 \pm 0.11$  in six homes in New Jersey, depending on ventilation rates and indoor gas combustion. K. Lee et al., 1999 measured I/O ozone ratios in 20 homes under different conditions, finding that the I/O ratio was 0.68 with air conditioning off and windows open, and significantly lower, at around 0.15, with windows and doors closed and an air cleaner on. Liu & Nazaroff, 2001b reported I/O ozone concentration ratios in 50 homes using passive ozone samplers, with a mean weekly I/O ratio of  $0.07 \pm 0.10$  in winter and  $0.40 \pm 0.29$  in summer. Cattaneo et al., 2011b reported an average I/O ozone concentration ratio of 0.22 in summer across 60 homes in Italy, while Brauner et al., 2014 found an average ratio of 0.05 in six elder care homes.

#### 4.4.- Ozone penetration factor

Significantly less research has focused on the penetration of ozone through building enclosure assemblies. In scenarios without indoor sources, steady-state estimates of I/O ratios can be calculated, substituting the penetration factor (P) with an ozone-specific value and replacing the deposition loss rate (k) with the first-order ozone decay rate. Weschler, 2006c noted that the penetration factor (P) is generally assumed to be equal to 1; however, this assumption has been largely untested in real buildings and has undergone only minimal testing in chamber studies.

#### 4.4.1.- Modeling studies

Liu & Nazaroff, 2001a modeled the penetration of reactive gases through idealized rectangular cracks and cavities filled with fiberglass insulation within building envelopes. Their findings indicated that penetration is influenced by both the geometry of the cracks and the reactivity of the crack surfaces, which is characterized by the material's reaction probability with the gas. The study predicted that ozone penetration through fiberglass insulation could range from over 90% to approximately 10 – 40%, depending on the reactivity of the fiberglass fibers. Walker & Sherman, 2013 extended the modeling methods of Liu & Nazaroff, 2001a to evaluate the effects of various ventilation systems on indoor ozone concentrations, concluding that mechanical ventilation systems used alongside typical building envelopes could reduce indoor ozone concentrations by 80 – 90% compared to outdoor levels.

#### 4.4.2.- Field measurements

Measurements of ozone penetration factors in real buildings are extremely limited. One notable study by Stephens & Siegel, 2012b developed a method to measure ozone penetration factors in residences. The test involved artificially elevating indoor ozone levels and monitoring the subsequent decay. A calibrated blower door fan was used to depressurize the test house, which increased the air exchange rate (AER) and elevated indoor ozone concentration above the detection threshold of a UV photometric ozone monitor. The ozone penetration factor was estimated along with the deposition loss rate constant by fitting data to a two-parameter model, while simultaneously measuring the AER with tracer gas injection and decay ( $\text{CO}_2$ ). This method was applied to eight single-family homes, yielding a mean ( $\pm$  SD) ozone penetration factor of  $0.79 \pm 0.13$ , with a range from  $0.62 \pm 0.09$  to  $1.02 \pm 0.15$ . However, the method's accuracy may be compromised, as the use of a blower door to increase AERs could have altered the characteristics of leakage pathways and the air velocity through leaks.

## 4.5.- Nitrogen oxides infiltration and penetration factors

The infiltration of outdoor NO<sub>x</sub> into indoor environments has been less extensively studied compared to particles and ozone, partly due to the influence of indoor sources such as cooking and space heating. López-Aparicio et al., 2011 monitored indoor and outdoor NO<sub>2</sub> levels for nine months in a naturally ventilated library, assuming no indoor sources – a reasonable assumption given the absence of a heating system in the building. The measured infiltration factor (i.e., I/O ratio) ranged from 0.21 to 0.33, with minor seasonal variations. Other studies conducted in residences likely included indoor sources. These studies employed the linear regression method to estimate the outdoor contribution to indoor NO<sub>x</sub> levels. For instance, Kulkarni & Patil, 2002 measured indoor and outdoor NO<sub>2</sub> concentrations in 43 houses in Mumbai, India, estimating mean infiltration factors of 0.92 and 0.73 for two types of households with different stoves. Similarly, W. Yang et al., 2004b carried out daily indoor and outdoor NO<sub>2</sub> measurements over 30 consecutive days in 28 houses in Brisbane, Australia, and over 21 consecutive days in 37 houses in Seoul, Korea, with mean estimated  $F_{inf}$  values of  $0.59 \pm 0.14$  in Brisbane and  $0.59 \pm 0.12$  in Seoul. Baxter et al., 2006b used a multilinear regression method, reporting a mean NO<sub>2</sub> infiltration factor of  $0.48 \pm 0.26$ . Wichmann et al., 2010 measured indoor and outdoor NO<sub>2</sub> levels during winter and summer in 18 homes, as well as in six schools and 10 pre-schools, finding that infiltration factors were influenced by the micro-environment, ventilation type and air exchange rate, with an aggregated value of 0.64. Meier et al., 2015 estimated I/O NO<sub>2</sub> concentration data from 66 homes, with the outdoor fractions of indoor NO<sub>2</sub> ranging from 37% to 61%, depending on window-opening frequency. Rivas et al., 2015b conducted two one-week sampling campaigns in 36 schools in Barcelona, finding that infiltration rates were affected by building age and window type, with an average value of 0.53.



## 5.- RESULTS

This chapter provides a summary of studies that have developed and refined methods to measure the transport of three major outdoor pollutants – ozone, particulate matter (PM) and nitrogen oxides (NO<sub>x</sub>) – into residential indoor environments, addressing the objectives outlined before. This chapter is organized into four sections, each corresponding to one of the research objectives.

### 5.1.- A refined method to measure the ozone penetration factor in residences under infiltration conditions

A refined test method was developed and tested in studio E, the Suite for Testing Urban Dwellings and their Indoor and Outdoor Environments. This unoccupied, sparsely furnished apartment unit is located on the third floor of a 9-story residence hall on the main campus of the Illinois Institute of Technology in Chicago, IL. The apartment unit has a floor area approximately 60 m<sup>2</sup> and a volume of around 150 m<sup>3</sup>. Approximately half of the perimeter walls are exterior walls, while the ceiling, floor and the other half of the perimeter walls are adjacent to other interior spaces, including other apartment units and the hallway. During the measurements, all windows and the perimeter door were kept closed, with internal doors kept open and several oscillating fans operated to enhance air mixing.

The test procedure was refined by integrating techniques from the only known previous ozone (O<sub>3</sub>) penetration test method with methods used in prior outdoor particle penetration measurements into homes ((Stephens et al., 2012); (Chao et al., 2003); (Thatcher et al., 2003)). The improved method utilized a manual indoor ozone elevation and decay procedure. Indoor ozone concentrations were first elevated to approximately 120 – 160 ppb using three UV ozone generators (CAP Model OZN – 1) placed in the bedrooms. A switching valve (Swagelok Model SS-43GXS4-42DCX) combined with an electronic timer (Sestos B3S-2R-24) was then used to alternate the ozone monitors between indoor and outdoor ozone concentration measurements at 3 – minute intervals (2 minutes indoors and 1 minute outdoors). Data points during the indoor/outdoor transition period were



excluded to ensure clear distinctions between indoor and outdoor sampling periods. The entire system was connected using PTFE – lined sampling lines to minimize transport loss. Air exchange rates during the test period were simultaneously measured using a CO<sub>2</sub> tracer gas injection and decay method.

### 5.1.1.- Example of data

The following figure presents an example of alternating indoor and outdoor ozone concentration data from two consecutive ozone injection and decay experiments conducted on the same day.

The indoor ozone first-order decay rate was initially calculated using a log-linear regression analysis applied to the early portion of the indoor decay data. With both the decay rate constant ( $k$ ) and air exchange rate (AER) determined, the ozone penetration factor was estimated using five different methods outlined beforehand: a steady-state solution, a one-parameter analytical solution (with  $k$  known), a two-parameter analytical solution, a one-parameter discretized solution (with  $k$  known) and a two-parameter discretized solution. Twenty-one replicate tests were conducted in the test apartment during the summer season under various indoor and outdoor conditions to assess the accuracy and repeatability of the refined natural infiltration test method and the efficacy of the different mathematical approaches for calculating ozone decay constants and penetration factors.

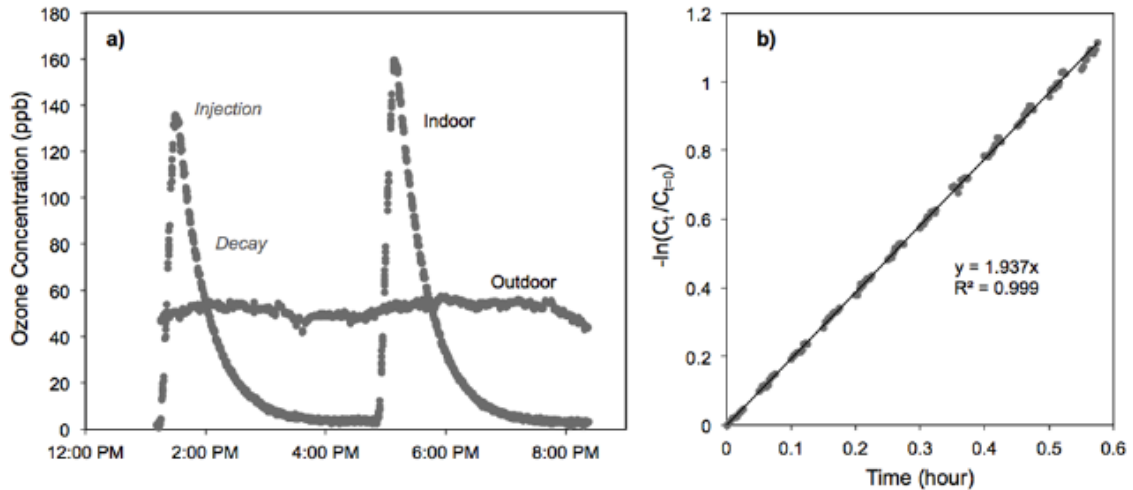


Figure 2.- Example of data from ozone injection and decay measurements: (a) timeseries data from two consecutive tests, and (b) solving for the ozone decay rate constant using log-linear regression.

### 5.1.2.- Instrument comparison

Indoor and outdoor ozone concentrations were measured using two UV absorbance ozone monitors:

1. 2B Technologies Model 205 dual-beam ozone monitor.
2. 2B Technologies Model 211 “interference-free” scrubberless ozone monitor.

Both monitors were connected to the same automated sampling system and operated simultaneously under identical air sampling conditions. While both monitors are listed on the US EPA’S designated federal equivalent methods for measuring ambient ozone concentrations (US EPA 2014), they differ in their method of operation and detection limits, which holds significant implications for indoor use.

The Model 205 ozone monitor utilizes a conventional solid-phase scrubber, with a reported accuracy of 1 ppb and a detection limit of 2 ppb. This monitor relies on UV absorption at 254 nm, a technology known to have interference issues with Hg, water vapor and various indoor VOCs, including styrene, methylstyrene, o-cresol, nitrocresol and other aromatic species with substituted electron-withdrawing groups (e.g., -OH, -NO<sub>2</sub> and -CHO) ((Ollison, Crow, et al., 2013); (Spicer et al., 2010); (Ollison, Capel, et al., 2013); (E. J. Williams et al., 2006); (Grosjean & Harrison,

1985b); (Huntzicker & Johnson, 1979); (Wilson & Birks, 2006); (H. Zhao & Stephens, 2016b)). In contrast, the “interference-free” Model 211 monitor, with a reported accuracy of 1 ppb and a detection limit of 1 ppb, employs a NO-O<sub>3</sub> gas-phase titration scrubber instead of a solid-phase scrubber, selectively removing O<sub>3</sub> while avoiding interference from other compounds that absorb UV light at the same wavelength.

The following figure presents a summary of the steady-state indoor and outdoor ozone concentrations measured by both ozone monitors during the 21 replicate tests conducted in the apartment unit under natural infiltration conditions.

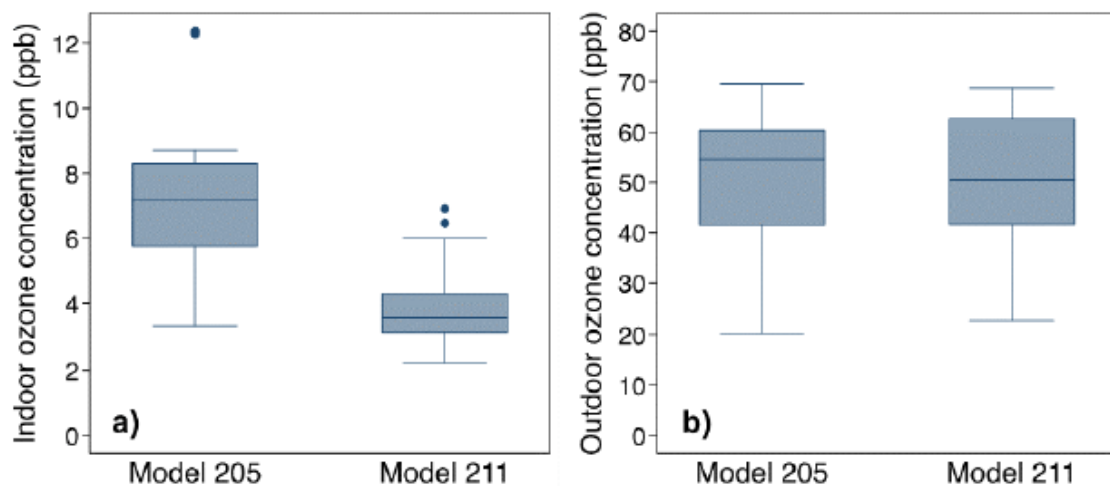


Figure 3.- Steady-state ozone concentrations were measured both indoors (a) and outdoors (b). The data is presented with boxes representing the 25th, 50th and 75th percentiles.

Both ozone monitors provided consistent outdoor ozone concentration measurements, with a mean ( $\pm$  SD) of  $51.2 \pm 12.9$  ppb for the Model 205 monitor and  $51.8 \pm 12.5$  ppb for the Model 211 monitor, ranging from 23 to 70 ppb across all tests. A Wilcoxon rank-sum test revealed no statistically significant difference between the two monitors’ outdoor measurements ( $p = 0.78$ ). However, the Model 205 monitor recorded significantly higher steady-state indoor ozone concentrations, with a mean ( $\pm$  SD) of  $7.2 \pm 2.4$  ppb, compared to  $4.0 \pm 1.3$  ppb from the Model 211 monitor. This resulted in higher indoor-to-outdoor (I/O) ozone concentration ratios for the Model 205, with a median of 0.14 compared to 0.09 ( $p = 0.0001$  according to a Wilcoxon rank-sum test). These findings indicate that the conventional UV

absorbance ozone monitor (Model 205) tended to overestimate indoor concentrations due to relatively high detection limits and/or interference at low indoor ozone concentrations. Given these implications for estimating penetration factors under natural infiltration conditions, the remainder of the work focuses solely on the results obtained from the Model 211 monitor to better demonstrate the utility of the new natural infiltration test method.

### 5.1.3.- Mathematical solution methods comparison

The mean estimates ( $\pm$  SD) of the ozone penetration factor (P) across all 21 natural infiltration tests in the test apartment were  $0.65 \pm 0.11$ ,  $0.47 \pm 0.08$ ,  $0.59 \pm 0.11$ ,  $0.54 \pm 0.10$  and  $0.55 \pm 0.12$ , depending on the method used: steady-state, one-parameter analytical, two-parameter analytical, one-parameter discretized and two-parameter discretized solutions, respectively. These results are shown below:

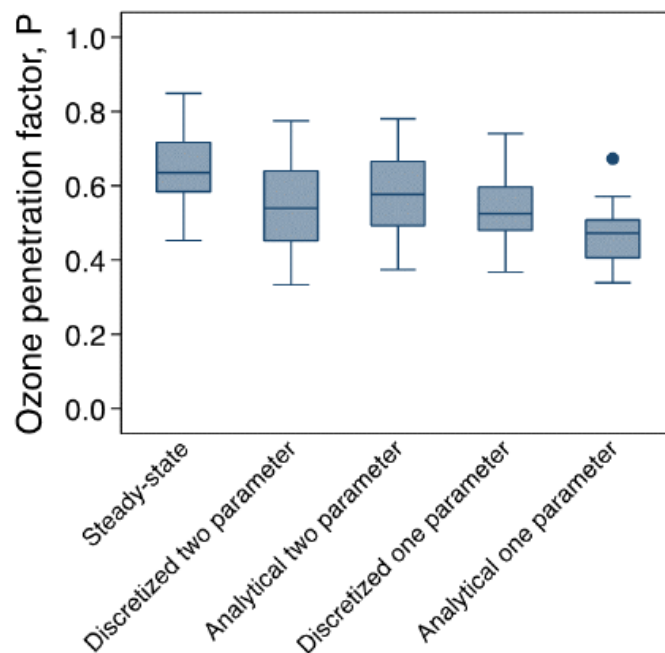


Figure 4.- Estimates of ozone penetration factors (P) were obtained across 21 replicate experiments conducted under natural infiltration conditions, utilizing five different solution methods with the Model 211 ozone monitor.

Estimates of the ozone penetration factor (P) were found to be relatively consistent across all solution methods, with particularly close alignment observed between the two discretized methods and the two-parameter analytical method. Median estimates of P varied, ranging from a maximum of 0.64 using the steady-state

solution method to a minimum of 0.48 using the analytical one-parameter method. The steady-state method yielded significantly higher estimates of P compared to other methods ( $P < 0.05$  for all comparisons except with the analytical two-parameter method, based on a Wilcoxon rank-sum test). Conversely, the analytical one-parameter method produced significantly lower estimates of P than all other methods (Wilcoxon rank-sum  $P < 0.05$ ). No significant differences were observed between the discretized one-parameter, discretized two-parameter and analytical two-parameter methods.

To further assess the model fit and accuracy of each solution method using data from the Model 211 monitor, two metrics were used:

1. Uncertainty estimates calculated through error propagation, combining relative uncertainties of the air exchange rate (AER) and the standard error of regression coefficients for P and k.
2. Mean squared errors (MSE).

The steady-state method exhibited the highest average uncertainty estimate at  $0.19 \pm 0.05$  largely due to the high relative standard deviations of the time-averaged indoor and outdoor ozone concentrations. The next highest uncertainty was observed with the two-parameter discretized solution method ( $0.13 \pm 0.04$ ). The one- and two-parameter analytical methods, along with the one-parameter discretized method, had similar uncertainty estimates:  $0.08 \pm 0.01$ ,  $0.09 \pm 0.02$  and  $0.09 \pm 0.03$ , respectively. These findings suggest that the analytical methods and the one-parameter discretized solution method are the most suitable for minimizing uncertainty.

Regarding model fit, the MSE was calculated for all 21 natural infiltration experiments using data from both analytical and discretized methods. The MSEs for the two analytical methods were higher than those for the two discretized methods, which were nearly identical. The average MSE for the one- and two-parameter analytical methods were  $0.26 \pm 0.23$  and  $0.15 \pm 0.16$ , respectively, compared to 0.10

$\pm 0.04$  for both the one- and two-parameter discretized method also offering the lowest estimated uncertainty.

Overall, the results indicate that measurements using the Model 211 ozone monitor, combined with parameter estimates from the one-parameter discretized solution method, provide the most consistent estimates of P and k under natural infiltration conditions, with the lowest combined uncertainty and MSE. This consistency likely stems from the discretized method's ability to account for varying outdoor concentrations while leveraging data from the initial log-linear decay to reliably estimate k.

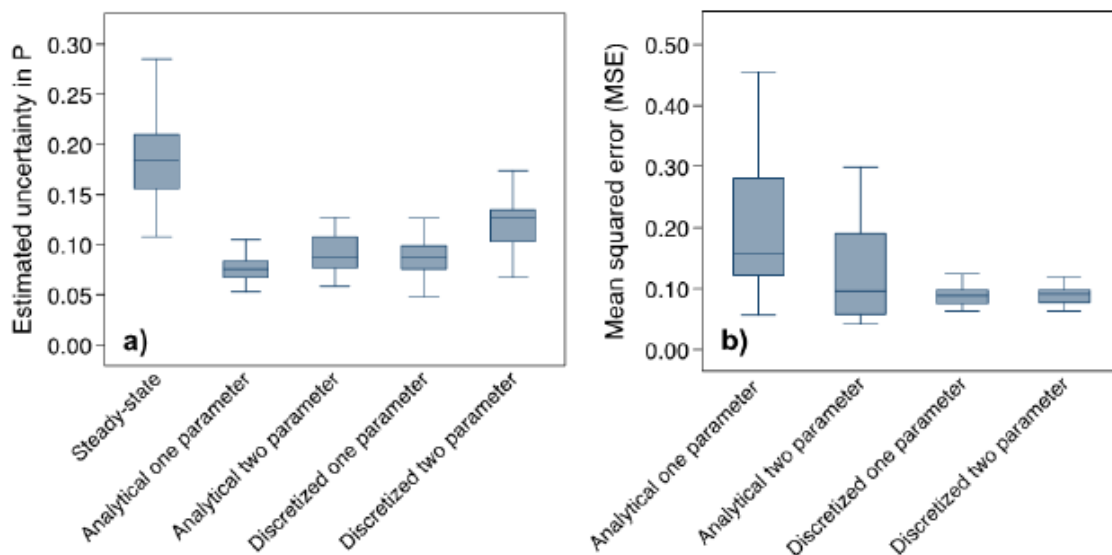


Figure 5.- Range of uncertainty estimates in 21 natural infiltration experiments using the Model 211 ozone monitor: (a) uncertainty in ozone penetration factor and (b) mean squared error.

#### 5.1.4.- Comparison to the original blower door test method

A subset of three tests was performed using the original fan depressurization test method as reported in Stephens & Siegel, 2012b. In these tests, a blower door was installed in the doorway to depressurize the unit relative to the hallway, elevating the air exchange rate ( $\lambda$ ) to approximately  $3 \text{ h}^{-1}$ . Both ozone monitors were used to collect data for comparison. The steady-state indoor ozone concentrations measured by both monitors are shown in Figure 6. The mean ( $\pm$  SD) indoor concentrations were similar:  $16.5 \pm 0.9$  ppb with the Model 211 monitor and  $17.9 \pm 1.8$  ppb with the Model 205 monitor, with no statistically significant difference ( $P = 0.28$  according to a

Wilcoxon signed-rank test). This suggests that the fan depressurization method successfully elevated indoor ozone concentrations above the detection limit of the Model 205 monitor.

Parameter estimates for  $\lambda$ ,  $k$  and  $P$  during the three blower door experiments were determined using three solution methods – steady-state, two-parameter analytical and one-parameter discretized – and compared to the mean ( $\pm$  SD) results from the 21 natural infiltration experiments. The data indicate that in this particular building, the blower door method tended to underestimate the ozone penetration factor relative to the values measured during natural infiltration across all three solution methods. However, the two-parameter analytical solution provided the closest agreement, suggesting that previous measurements of the penetration factor ( $P$ ) reported in Stephens & Siegel, 2012b may still be considered reasonable estimates.

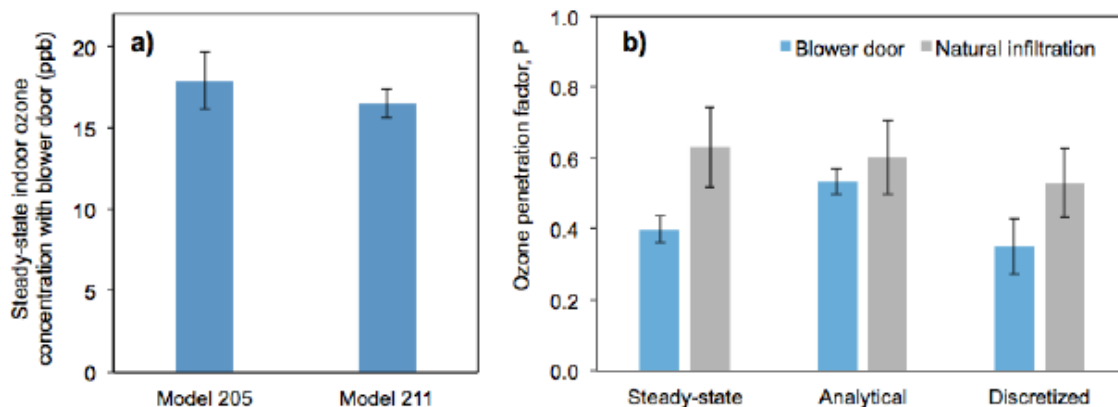


Figure 6.- Comparison of natural infiltration results with the blower door test. (a) steady-state indoor ozone concentrations measured by both ozone monitors, (b) comparison of ozone penetration factor ( $P$ ).

## 5.2.- An improved method to rapidly measure the penetration of fine and ultrafine particles in unoccupied residences

The particle penetration test method was primarily developed based on the principles of the novel ozone penetration method described earlier, along with other established methods in the existing literature. The method was developed and tested in studio E using a natural elevation and decay procedure. Given prior experience, only the discretized method was used to solve for the penetration factor



(P) and the decay constant ( $k$ ), with  $k$  being determined in advance using only the decay data.

The test utilized a combination of instruments: a TSI NanoScan Scanning Mobility Particle Sizer (SMPS, TSI NanoScan Model 3910) and a TSI Optical Particle Sizer (OPS, TSI Model 3330), which measured particle concentrations ranging from  $\sim 10$  nm to  $\sim 10$   $\mu$ m in mobility and optical diameter, respectively. The system also included DustTrak DRX aerosol monitor (TSI DRX Model 8534), capable of measuring size-segregated mass fraction concentrations corresponding to PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> size fractions using optical methods. All three portable instruments were connected to the same sampling system using TSI conductive silicon tubing and placed in a medium-sized audio rack case on rollers, located centrally in the living room adjacent to an exterior window. Two grid stainless-steel sampling lines, approximately 1.5 meters in length and 0.6 cm in diameter, were used for both indoor and outdoor sampling to minimize particle deposition losses and ensure similar loss rates for both sampling lines. The entire sampling system was well-sealed, with total transport losses consistently measured at approximately 15%.

Outdoor particles were introduced by opening windows and operating a blower door fan installed in a doorway to induce cross-ventilation flow. This procedure elevated indoor particle concentrations to levels close to outdoor concentrations and replaced indoor aerosols with those matching the outdoor composition. The unit was then left unoccupied to measure the subsequent decay of indoor particles with all exterior windows and doors closed. Indoor particle measurements continued for 40 – 60 minutes during an indoor-only measurement period prior to initiating indoor/outdoor switching. After this initial decay period, indoor and outdoor particle concentrations were alternately measured at 8-minute intervals (4 minutes indoors and 4 minutes outdoors) using a similar switching system as the one employed in the ozone penetration test method. These alternating measurements enabled tracking of changes in indoor pollutant concentrations due to outdoor infiltration.

The air exchange rate during the test period was measured using CO<sub>2</sub> injection and decay, following the same approach as the ozone penetration method.

In addition to the size-resolved data (which in some size bins had concentrations too low to yield meaningful elevation and decay signatures), integral measures of indoor and outdoor ultrafine particle (UFP) number concentrations were calculated at each time step by summing concentrations from the first 8 bins of the SMPS (i.e., 10 nm to 101 nm). Similarly, integral measures of indoor and outdoor PM<sub>2.5</sub> mass concentrations were estimated at each time step by calculating the mass concentration in each particle size bin smaller than 2.5 µm using data from the SMPS and OPS, assuming spherical particle shape and two different density conditions:

1. Constant unit density.
2. Density varying with diameter.

A total of 11 replicate tests were conducted in the test apartment under a wide range of indoor and outdoor environmental conditions to assess the repeatability of the method.

### 5.2.1.- Example of data

An example of resulting indoor and outdoor integral ultrafine particle (UFP) concentration data from a typical elevation and decay experiment is shown in figure 7a. The initial portion of the indoor decay data was used to determine the UFP deposition loss rate constant ( $k$ ) for this experiment, as illustrated in figure 7b. The air exchange rate (AER) during this test was  $1.16 \pm 0.05/h$ , with the estimated value for  $k$  being  $0.89 \pm 0.09/h$ . Subsequently, size-resolved particle penetration factors were calculated.

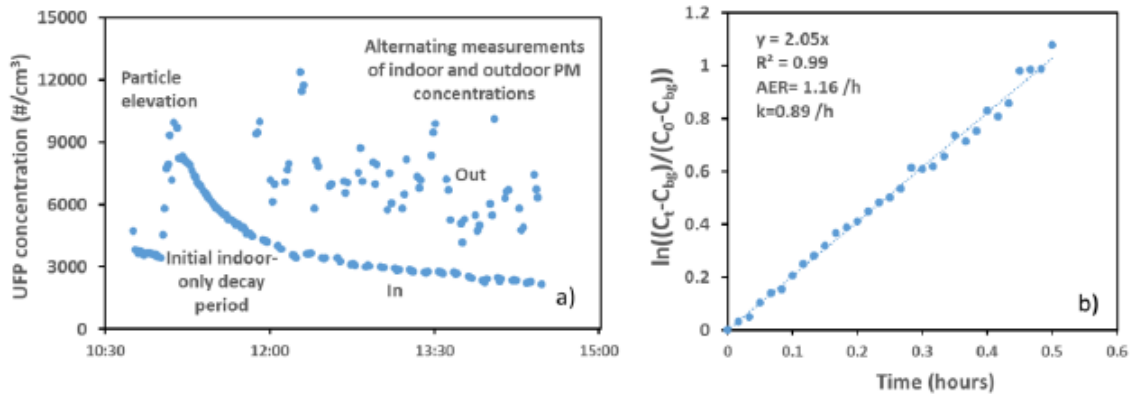


Figure 7.- Example of data from a particle elevation and decay measurement. (a) time-series integral UFP data from one test, (b) determination of the integral UFP deposition loss rate constant using a log-linear regression solution.

### 5.2.2.-Instrument comparison

Results for estimates of infiltration factors for integral measures of UFPs and PM<sub>2.5</sub> based on the underlying size-resolved infiltration factors for the 11 replicate tests are presented in figure 8, along with PM<sub>2.5</sub> infiltration factors measured using the TSI DustTrak during the same periods. The mean ( $\pm$  SD) infiltration factors for UFPs, PM<sub>2.5</sub> estimated assuming constant unit density, PM<sub>2.5</sub> estimated assuming varying density, and PM<sub>2.5</sub> measured with the DustTrak, averaged over all 11 replicate tests, were  $0.39 \pm 0.05$ ,  $0.49 \pm 0.09$ ,  $0.49 \pm 0.08$  and  $0.49 \pm 0.18$ , respectively. The infiltration factors for UFPs were relatively consistent. The infiltration factors for PM<sub>2.5</sub> estimated using the two different density assumptions did not differ significantly, indicating that the PM<sub>2.5</sub> infiltration factor estimates using the SMPS + OPS system were not sensitive to variations in particle density assumptions. The mean and median PM<sub>2.5</sub> infiltration factors measured using the DustTrak were slightly lower than those estimated with the SMPS + OPS combination under either density assumption, but these were more variable and sometimes yielded unreasonable values due to low indoor concentrations.

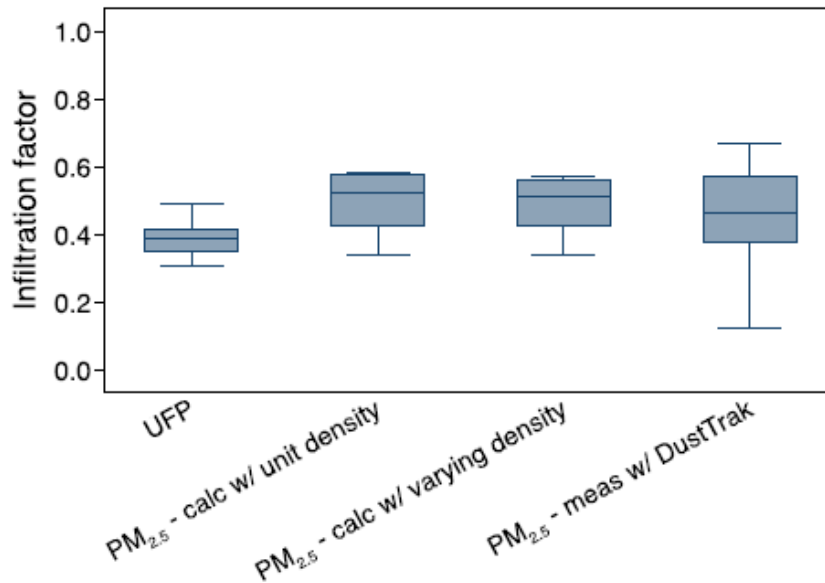


Figure 8.- Infiltration factors estimated for integral measures of UFPs, mass concentrations of particle less than 2.5  $\mu\text{m}$  assuming unit density, varying density and PM<sub>2.5</sub> measured with the TSI DustTrak.

Figure 9a illustrates that the infiltration factor ( $F_{\text{inf}}$ ) values for PM<sub>2.5</sub> estimated using data from the SMPS + OPS measuring system, applied with two different density assumptions, did not differ significantly. Figure 8b compares the PM<sub>2.5</sub> infiltration factors estimated using the SMPS + OPS system with the constant density assumption to those measured using the DustTrak. A strong correlation was observed between the PM<sub>2.5</sub> infiltration factors measured by the DustTrak and the SMPS + OPS system ( $R^2 = 0.91$ ), indicating that the SMPS + OPS system provides reasonable estimates of  $F_{\text{inf}}$  for PM<sub>2.5</sub>. However, the DustTrak generally underestimated PM<sub>2.5</sub> infiltration factors compared to the SMPS + OPS system. This discrepancy may be attributed to the DustTrak's reliance on optical measurements to infer PM<sub>2.5</sub> mass concentrations, which do not account for particles below 0.1  $\mu\text{m}$  (as specified in the manufacturer's product literature) or potentially even higher sizes (e.g., below approximately 0.3  $\mu\text{m}$  for most optical instruments). Depending on the outdoor particle size distribution, a significant portion of PM<sub>2.5</sub> mass may exist below the 0.1 or 0.3  $\mu\text{m}$  threshold, which the DustTrak does not capture, whereas the SMPS + OPS system is capable of detecting these smaller particles.

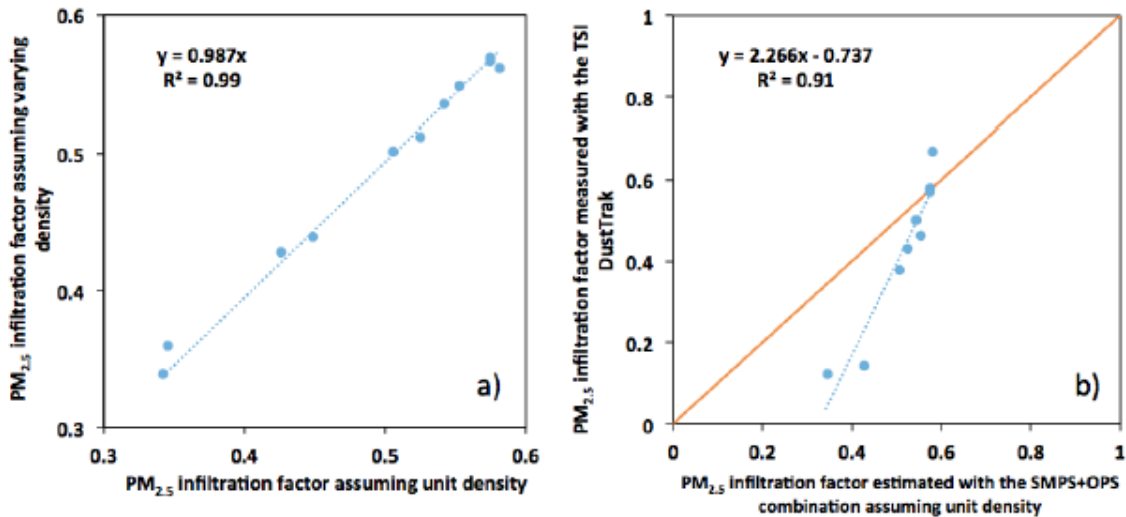


Figure 9.- Regression analysis between (a) PM<sub>2.5</sub> infiltration factors calculated using two different density assumptions, (b) PM<sub>2.5</sub> infiltration factors calculated assuming constant unit density and measured using a TSI DustTrak.

### 5.2.3.- Size – resolved particle infiltration factors, penetration factors and deposition loss rate constants

Figure 10 illustrates the size-resolved steady-state indoor and outdoor particle infiltration factors, measured by both the SMPS and OPS, across 18 size bins ranging from 0.01  $\mu\text{m}$  to 2.5  $\mu\text{m}$  in mobility and optical diameter, respectively. These measurements were taken during each of the 11 replicate tests conducted in the test apartment unit under infiltration conditions. The thick black line represents the average value of the infiltration factor ( $F_{\text{inf}}$ ) across all 11 tests. The air exchange rates, which varied from 0.49  $\text{h}^{-1}$  to 1.16  $\text{h}^{-1}$  during each test period, are also provided in the legend. Additionally, the same data were applied to derive size-resolved penetration factors (Figure 11a) and deposition loss rate constants (Figure 11b) for the first 13 particle size bins ranging from 0.01  $\mu\text{m}$  to 1  $\mu\text{m}$  were insufficient (and exhibited too much scatter) to yield reliable estimates of the deposition loss rate constant ( $k$ ) for those bins ( $R^2$  was consistently below 0.5). However, the steady-state indoor/outdoor (I/O) concentrations in these larger bins were adequate to calculate  $F_{\text{inf}}$ , as shown in Figure 10.

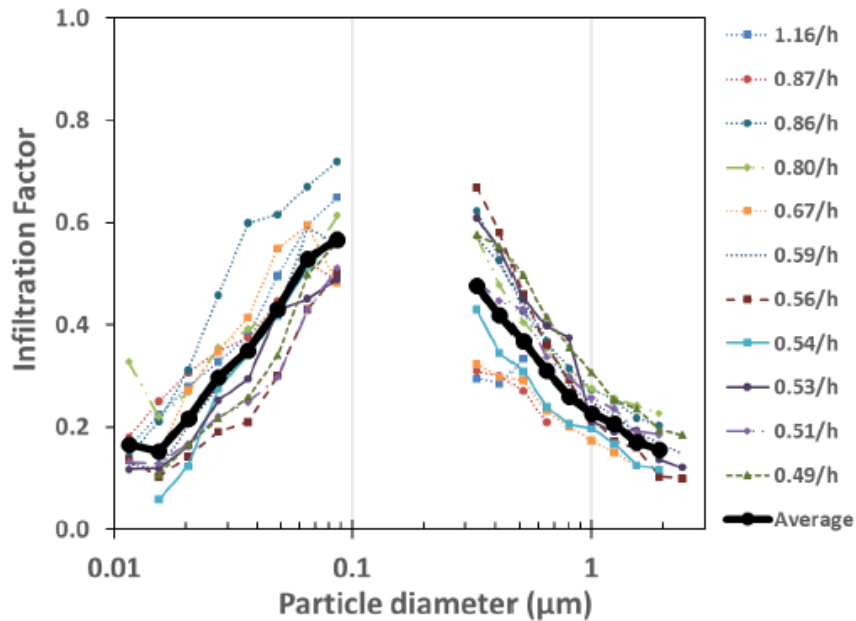


Figure 10.- Size-resolved particle infiltration factors with particle diameters ranging from 0.01  $\mu\text{m}$  to 2.5  $\mu\text{m}$  in mobility and optical diameter, respectively, with various AER in the apartment unit.

Size-resolved envelope penetration factors generally increased with particle size for ultrafine particles (UFPs) and decreased for particles larger than 0.3  $\mu\text{m}$ . This pattern aligns with particle penetration theory, where larger particles are more likely to be removed by gravitational settling cracks, while smaller particles are more susceptible to removal by Brownian diffusion ((Liu & Nazaroff, 2001a); (Liu & Nazaroff, 2003b)). Across the 11 replicate tests, the average ( $\pm$  standard deviation) penetration factors ranged from a minimum of  $0.41 \pm 0.14$  at a geometric mean particle diameter of 15 nm to a maximum of  $0.73 \pm 0.05$  at 87 nm (Figure 11a). These findings are consistent with the limited prior measurements of size-resolved penetration factors reported in the literature C. Chen & Zhao, 2011. Similarly, the size-resolved deposition loss rate constants followed a characteristic U-shaped curve, generally decreasing with increasing particle size for UFPs and increasing for particles larger than 0.3  $\mu\text{m}$ . In this sparsely furnished apartment unit, with no central HVAC systems operating, the mean ( $\pm$  standard deviation) deposition loss rate constants ranged from a maximum of  $1.58 \pm 0.34 \text{ h}^{-1}$  for 11 nm particles to a minimum of  $0.11 \pm 0.07 \text{ h}^{-1}$  for 86 nm particles (Figure 11b).

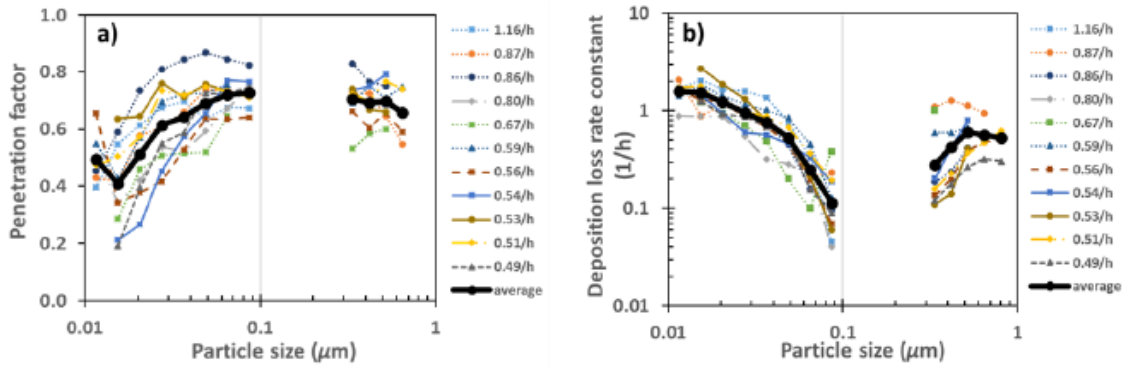


Figure 11.- Size-resolved (a) penetration factors, (b) deposition loss rate constants estimated across 11 replicate tests with varying AER.

#### 5.2.4.- Integral UFP and PM<sub>2.5</sub> penetration factors (P) and deposition loss rate (k)

Figure 12 presents the distributions of the estimated (a) penetration factors (P) and (b) deposition loss rate constants (k) for integral measures of ultrafine particles (UFPs) and PM<sub>2.5</sub> across the 11 replicate tests conducted in the apartment unit. The distributions also include estimates of their associated uncertainties. The mean ( $\pm$  standard deviation) estimates of P were  $0.67 \pm 0.05$  for UFPs and  $0.73 \pm 0.05$  for PM<sub>2.5</sub>, with ranges of 0.59 to 0.78 for UFPs and 0.65 to 0.79 for PM<sub>2.5</sub>. Estimates of k exhibited greater variability, with mean ( $\pm$  standard deviation) values of  $0.68 \pm 0.27$  h<sup>-1</sup> for UFPs and  $0.45 \pm 0.30$  h<sup>-1</sup> for PM<sub>2.5</sub>. The mean ( $\pm$  standard deviation) relative uncertainty in P for PM<sub>2.5</sub> and UFPs across the 11 replicate tests was  $12 \pm 5\%$  and  $11 \pm 4\%$ , respectively. Similarly, the relative uncertainty in k was  $12 \pm 7\%$  for PM<sub>2.5</sub> and  $13 \pm 6\%$  for UFPs.

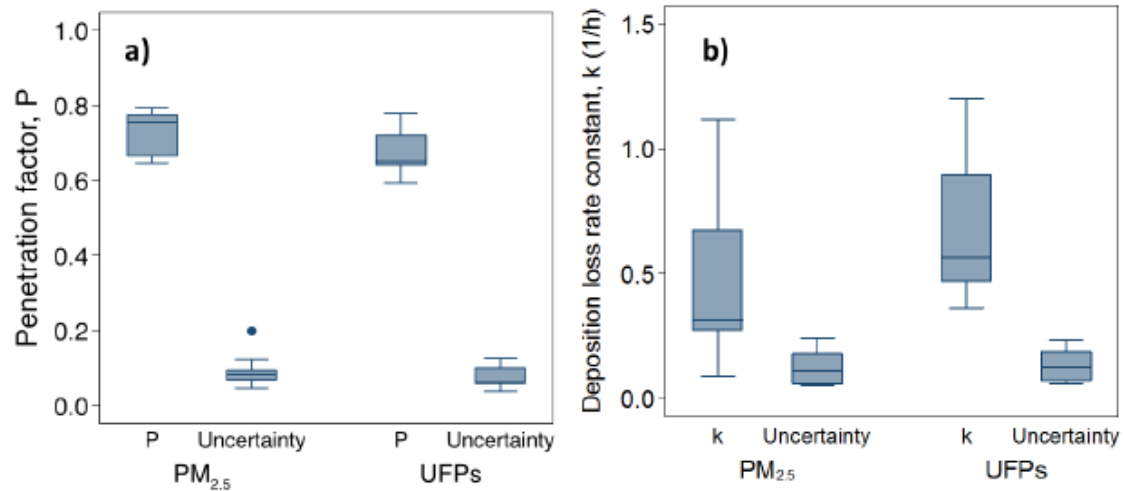


Figure 12.- Estimates of (a) penetration factors and (b) deposition loss rate constants for integral measures of UFPs and mass concentrations of PM<sub>2.5</sub> with associated uncertainties.

### 5.2.5.- Influence of indoor and outdoor environmental factors on estimates of P and k

The results of the estimates for penetration factors (P) and deposition loss rate constants (k), along with their associated uncertainties for integral measures of ultrafine particles (UFPs) and PM<sub>2.5</sub> from all 11 replicate penetration tests, were analyzed in relation to various coincident indoor and outdoor environmental conditions. These conditions included data on indoor and outdoor temperature, relative humidity, wind speed and wind direction. Spearman's rank correlation analysis between P and all indoor and outdoor environmental conditions revealed no significant correlations ( $p > 0.01$ ). This suggests that the test procedure and solution methods proposed for estimating the penetration factors for both UFPs and PM<sub>2.5</sub> were not significantly influenced by the observed ranges of environmental conditions. However, the estimates of the deposition loss rate constants (k) for both UFPs and PM<sub>2.5</sub> were significantly and negatively correlated with the geometric mean diameter of the average outdoor particle size distribution. The Spearman's rank correlation coefficients were -0.74 ( $p = 0.010$ ) for UFPs and -0.85 ( $p = 0.008$ ) for PM<sub>2.5</sub>. This outcome aligns with deposition theory, as outdoor particle size distributions with a larger geometric mean diameter (i.e., closer to 0.1  $\mu\text{m}$  rather than 0.01  $\mu\text{m}$ ) are expected to have lower deposition loss rate constants.



### 5.3.- Novel methods to measure the penetration factor of nitrogen oxides

The nitrogen oxides (NO<sub>x</sub>) penetration measurements were conducted in the same test apartment unit as described in previous sections. Indoor and outdoor concentrations of NO<sub>2</sub>, NO and total NO<sub>x</sub> (NO + NO<sub>2</sub>) were monitored using a 2B Technologies Model 405 direct UV-absorbance monitor, which logged data at 1-minute intervals. The instrument measured NO<sub>2</sub> directly via absorbance at 405 nm, while NO was measured by sequential conversion to NO<sub>2</sub> using internally generated ozone. The total NO<sub>x</sub> concentration was calculated by summing the measured NO and NO<sub>2</sub> concentrations. A switching system, similar to that described earlier, was used to alternate between indoor and outdoor NO<sub>x</sub> monitoring. The air exchange rates (AER) during these tests were periodically measured using the same CO<sub>2</sub> injection and decay method previously employed.

Initial short-term tests (4 – 6 hours), similar to those used for measuring ozone and particle penetration factors, were attempted. However, these efforts were unsuccessful due to slow instrument response times and intermittent low ambient and indoor NO<sub>x</sub> concentrations, which hindered accurate estimation of the penetration factor (P) and deposition loss rate constant (k). As a result longer-term measurements (approximately 24 – 48 hours) were adopted for the remainder of the testing. These longer tests used two primary experimental approaches, with 12 successful tests conducted using either method.

#### *Method 1: natural elevation and decay*

In this approach, indoor and outdoor NO and NO<sub>2</sub> concentrations were alternately measured at 40-minute intervals (20 minutes indoors and 20 minutes outdoors) over a period of at least 24 hours, with no indoor sources present, to capture the natural variations in indoor NO<sub>x</sub> concentrations due to changes in ambient NO<sub>x</sub> levels and infiltration rates into the apartment unit. To estimate the two unknowns, the penetration factor (P) and the deposition loss rate constant (k), a non-linear two-

parameter least squares regression was applied to the discretized solution of the indoor dynamic mass balance for total NO<sub>x</sub> concentrations:

$$C_{inNOx,t} = P_{NOx} \lambda_t C_{outNOx,t} \Delta t + (1 - (\lambda_t + k_{NOx}) \Delta t) C_{inNOx,t-1}$$

Equation 6.

Using the estimated total NO<sub>x</sub> penetration factor, the NO<sub>2</sub> penetration factor was calculated assuming that the NO penetration factor was equal to 1, based on the lack of literature indicating significant reactions between NO and common building materials. The NO<sub>2</sub> penetration factor was then determined by applying the estimated P for NO<sub>x</sub>, weighted by the average outdoor NO<sub>2</sub> and NO<sub>x</sub> concentrations during the test period, and assuming P<sub>NO</sub> = 1, as shown:

$$P_{NO2} * \overline{C_{outNO2}} + P_{NO} * \overline{C_{outNO}} = P_{NOx} * \overline{C_{outNOx}}$$

Equation 7.

Method 1 yielded only moderate success, primarily because natural peaks and decays in indoor NO<sub>x</sub> concentrations were not large enough to successfully apply the two-parameter regression fit. As a result, the test procedure was revised to include an artificial elevation and decay approach (Method 2), which aimed to provide more detailed insight into the behavior of individual NO<sub>x</sub> constituents under a wider range of conditions.

#### *Method 2: artificial elevation and decay*

In Method 2, the test procedure was modified to include a short-term artificial elevation and decay of NO<sub>2</sub> and NO concentrations, either before or after the longer-term natural elevation and decay measurements. Indoor NO<sub>2</sub> and NO levels were artificially elevated by operating a portable butane gas stove in one of the bedrooms for approximately 15 minutes. Since the gas stove produced significantly more NO than NO<sub>2</sub>, an ozone generator (CAP Model OZN – 1) was used concurrently with the stove to enhance the conversion of NO to NO<sub>2</sub> through the reaction between NO and O<sub>3</sub>. This typically resulted in a peak concentration of around 50 – 100 ppb.

During the test, the NO<sub>x</sub> monitor was programmed to measure indoor NO/NO<sub>2</sub> concentrations for the first two hours following the elevation. The indoor decay rates of NO<sub>2</sub> and NO during the ~ 2-hour decay period were estimated using log-linear regression of the indoor concentration (adjusted for background levels) over time. Subsequently, the NO<sub>2</sub> penetration factor was calculated using two different approaches.

### Method 2a

Method 2a was applied to estimate parameters when the average indoor NO concentration measured during the test closely matched the average outdoor NO concentration. In such cases, it was assumed that indoor oxidative reactions involving NO were negligible (i.e.,  $k_{NO}$  should be approximately 0 h<sup>-1</sup>). This assumption could later be verified through an alternative approach. Furthermore, since  $G_{NO_2}$  is primarily due to reactions between NO and oxidants like ozone, hydroperoxyl radicals and alkylperoxy radicals, when  $k_{NO}$  was near zero, it was assumed that  $G_{NO_2}$  was also negligible.

Thus, the penetration factor for NO<sub>2</sub> ( $P_{NO_2}$ ) could be estimated using a one-parameter regression on the discretized solution for NO<sub>2</sub> concentrations, where  $k_{NO_2}$  was determined from the decay period, and  $G_{NO_2}$  was assumed to be zero.

$$C_{in,NO_2,t} = P_{NO_2} \lambda C_{out,NO_2,t} \Delta t + (1 - (\lambda t + k_{NO_2}) \Delta t) C_{in,NO_2,t-1} + G_{NO_2} \Delta t$$

*Equation 8.*

### Method 2b

Method 2b was employed when the average indoor NO concentration during the test period was lower than the average outdoor NO concentration. In such cases, low indoor-to-outdoor NO concentration ratios were assumed to indicate the occurrence of indoor oxidative reactions between NO and certain (though unmeasured) indoor oxidants, such as ozone, hydroxyl radicals (OH), hydroperoxyl radicals (HO<sub>2</sub>), or alkylperoxy radicals (RO<sub>2</sub>). The time-averaged indoor NO loss rate ( $k_{NO}$ ) for the test period was estimated using a one-parameter regression equation, with the assumption that  $P_{NO} = 1$ .

In these cases, the indoor NO<sub>2</sub> generation rate at each time step was assumed to equal the indoor NO consumption rate, based on the simplifying assumption that NO oxidation reactions produced NO<sub>2</sub> in a 1:1 ratio. Once the air exchange rate (AER), k<sub>NO2</sub> and G<sub>NO2</sub> were known, P<sub>NO2</sub> could be solved using:

$$C_{inNO,t} = P_{NO} \lambda C_{outNO,t} \Delta t + (1 - (\lambda_t + k_{NO}) \Delta t) C_{inNO,t-1}$$

Equation 9.

$$G_{NO2} = k_{NO} C_{inNO,t-1}$$

Equation 10.

### 5.3.1.- Example test data

An example of the resulting data from a single artificial elevation and decay experiment, followed by approximately 24 hours of alternating indoor and outdoor measurements without indoor sources, is shown in the figure below. During the decay period, the air exchange rate (AER) was 0.48 ± 0.02 h<sup>-1</sup>, ranging from 0.31 to 0.42 h<sup>-1</sup> during the subsequent 24 hours of natural indoor and outdoor monitoring. The estimates of k<sub>NO2</sub> and k<sub>NO</sub> were derived from the decay period data using log-linear regression, yielding values of 0.35 ± 0.03 h<sup>-1</sup> and 0.04 ± 0.03 h<sup>-1</sup>, respectively.

Since the estimate of k<sub>NO2</sub> was very close to zero and the average indoor and outdoor NO concentrations during the subsequent 24-hour period were nearly identical (with averages of 24.8 ppb and 24.4 ppb, respectively), Method 2a's criteria were applied. This assumed that indoor oxidative reactions with NO were negligible (setting k<sub>NO</sub> = 0) and thus G<sub>NO2</sub> was also assumed to be zero. Based on these assumptions, the penetration factor for NO<sub>2</sub> (P<sub>NO2</sub>) was estimated to be 0.78 ± 0.11 using one-parameter regression, incorporating prior estimates of k<sub>NO2</sub> and AER over time.

For comparison, Method 1 was also applied to solve for P<sub>NOx</sub>, using a two-parameter solution, based on the 24-hour indoor and outdoor measurements without the artificial decay period. This method yielded estimates of P<sub>NOx</sub> = 0.89 ± 0.10 and k<sub>NOx</sub> = 0.05 ± 0.02. Using the assumption of P<sub>NO</sub> = 1 and average ambient NO<sub>2</sub> and NO

concentrations of 19.8 ppb and 24.4 ppb, respectively, during the testing period,  $P_{NO_2}$  was calculated to be  $0.74 \pm 0.10$ , which was within approximately 5% of the result from Method 2a.

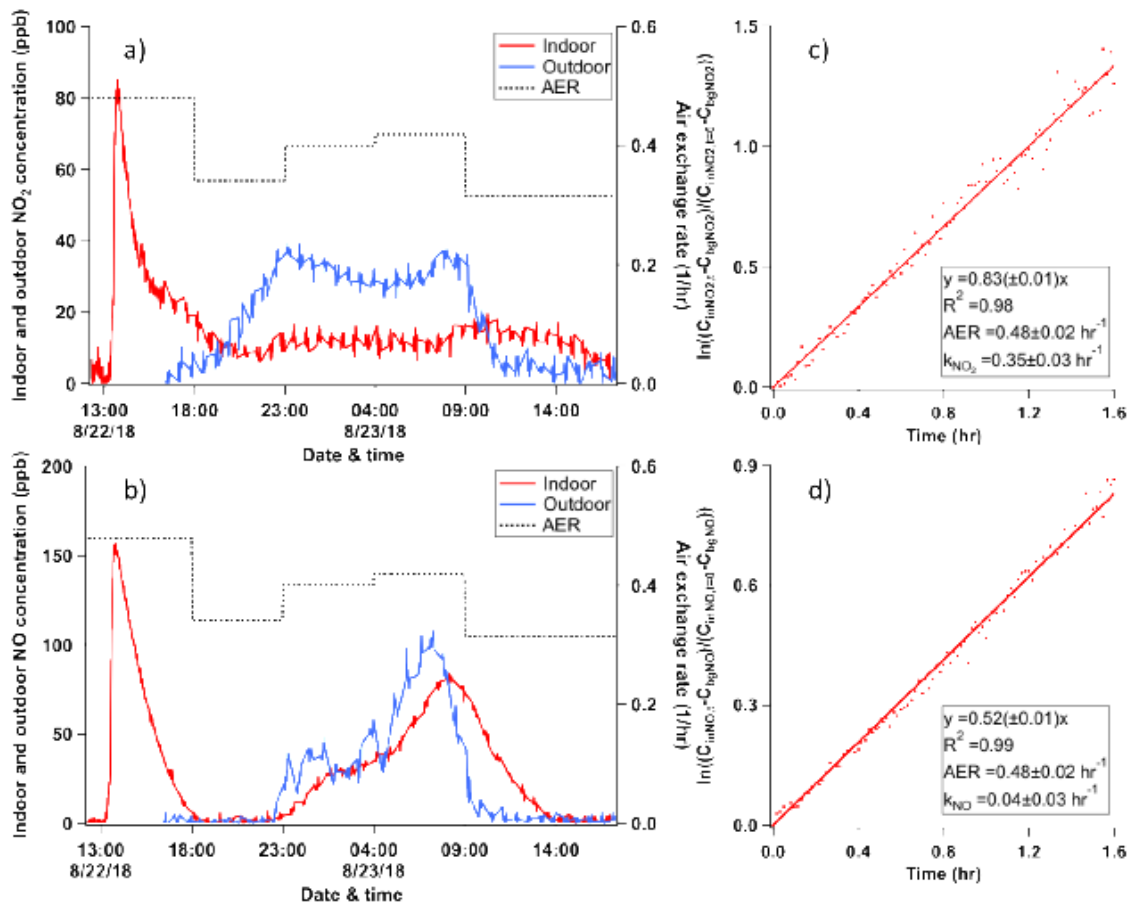


Figure 13.- Example test data from a single  $NO_x$  elevation and decay period, followed by natural variations, with parameters estimated successfully using both Method 1 and Method 2a.

Another example of data collected from a 24-hour period of alternating indoor and outdoor concentration measurements, followed by a single artificial elevation and decay experiment, is presented in Figure 15. During the decay period, the air exchange rate (AER) was  $1.14 \pm 0.01 \text{ h}^{-1}$ , while it ranged from 0.53 to  $0.82 \text{ h}^{-1}$  during the remaining  $\sim 24$ -hour monitoring period under natural conditions. Estimates of  $k_{NO_2}$  and  $k_{NO}$  were again derived from the decay period, resulting in values of  $0.18 \pm 0.12 \text{ h}^{-1}$  and  $0.25 \pm 0.09 \text{ h}^{-1}$ , respectively. As  $k_{NO}$  was greater than zero and the average indoor NO concentration during the 24-hour period was lower than the outdoor concentration (1.8 ppb versus 4.2 ppb), the decision criteria outlined in Method 2b were applied. This assumed that indoor oxidative reactions involving NO were

significant, and  $G_{NO_2}$  at each time step was estimated. Based on these assumptions,  $P_{NO_2}$  was determined to be  $0.73 \pm 0.10$  for this test case, using one-parameter regression equation and previous estimates of  $k_{NO_2}$  and  $k_{NOx}$  from the data (i.e.,  $P_{NOx} < 1$  and  $k_{NOx} > 0h^{-1}$ ). However, using the average outdoor NO and  $NO_2$  concentrations during the test period (4.2 and 10.7 ppb, respectively) and assuming  $P_{NO} = 1$ ,  $P_{NOx}$  was estimated to be  $0.80 \pm 0.15$ .

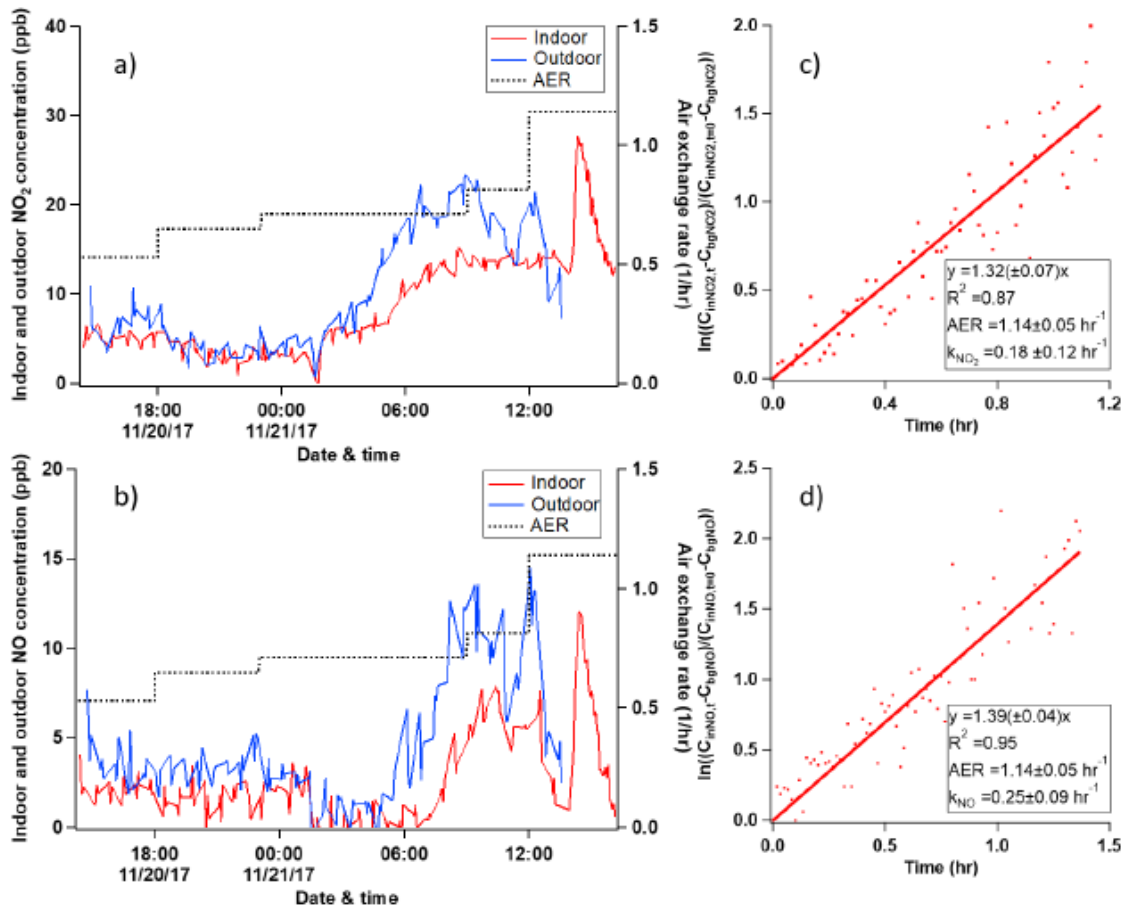


Figure 14.- Example test data from a single  $NOx$  elevation and decay period followed by natural variations, with parameters estimated using Method 1 (fail) and Method 2b (success).

Figure 15 presents the distributions of parameter estimates and associated uncertainties for both the  $NO_2$  penetration factor ( $P_{NO_2}$ ) and the indoor loss rate ( $k_{NO_2}$ ) based on the 12 successful tests, which were analyzed using either Method 1, Method 2a or Method b as appropriate. The mean ( $\pm$  S.D.) estimates of  $P_{NO_2}$  derived from the natural elevation and decay approach (Method 1,  $n = 7$ ), artificial elevation without indoor  $NO_2$  generation via NO oxidation (Method 2a,  $n = 7$ ), and artificial elevation with indoor  $NO_2$  generation via NO oxidation (Method 2b,  $n = 5$ ) were 0.69

$\pm 0.05$ ,  $0.72 \pm 0.05$  and  $0.76 \pm 0.07$ , respectively. These values ranged from 0.61 to 0.88, with an overall mean ( $\pm$  S.D.) of  $0.72 \pm 0.06$ . This suggests that approximately 28% of ambient  $\text{NO}_2$  was removed as it infiltrated the building envelope, indicating that reactions within the envelope reduced ambient  $\text{NO}_2$  exposure by an average of  $\sim 28\%$ .

A Wilcoxon rank-sum test revealed no statistically significant differences ( $p > 0.05$ ) between the  $P_{\text{NO}_2}$  estimates derived from Method 1 and Method 2 (grouping 2a and 2b together) or when Methods 1, 2a and 2b were treated separately. The relative uncertainty in  $P_{\text{NO}_2}$  was estimated to be  $15 \pm 2\%$ ,  $14 \pm 1\%$  and  $17 \pm 4\%$  for Methods 1, 2a and 2b, respectively, which is similar to previously reported uncertainties for ozone and particle penetration factors.

Estimates of  $k_{\text{NO}_2}$  were more widely distributed across the 12 tests, irrespective of the method used (Figure 15b). The mean ( $\pm$  S.D.) estimates of  $k_{\text{NO}_2}$  were  $0.27 \pm 0.10 \text{ h}^{-1}$  (Method 1),  $0.30 \pm 0.11 \text{ h}^{-1}$  (Method 2a), and  $0.22 \pm 0.13 \text{ h}^{-1}$  (Method 2b), with values ranging from 0.06 to  $0.47 \text{ h}^{-1}$  and an overall mean ( $\pm$  S.D.) of  $0.27 \pm 0.12 \text{ h}^{-1}$ . The  $k_{\text{NO}_2}$  values observed in this study were somewhat lower than those reported in previous studies conducted in real homes but were similar to values measured in a home in southern California. The lower  $k_{\text{NO}_2}$  values may be attributed to the sparsely furnished test apartment, which had a low surface-area-to-volume ratio, and the winter testing conditions with low relative humidity (RH), which is known to influence  $k_{\text{NO}_2}$ .

The relative uncertainty in  $k_{\text{NO}_2}$  was  $16 \pm 8\%$ ,  $28 \pm 25\%$  and  $52 \pm 33\%$  for Methods 1, 2a and 2b, respectively. The higher uncertainty in Method 2b was primarily due to the lower  $k_{\text{NO}_2}$  values under these conditions, though absolute uncertainties were similar across all methods. The Wilcoxon rank-sum test indicated no significant differences in  $k_{\text{NO}_2}$  estimates across the three methods ( $p > 0.05$ ). The overall consistency in  $P$  and  $k$  estimates across methods suggests that the approaches used here effectively capture the behavior of  $\text{NO}_x$  constituents, especially  $\text{NO}_2$  under a wide range of test conditions.

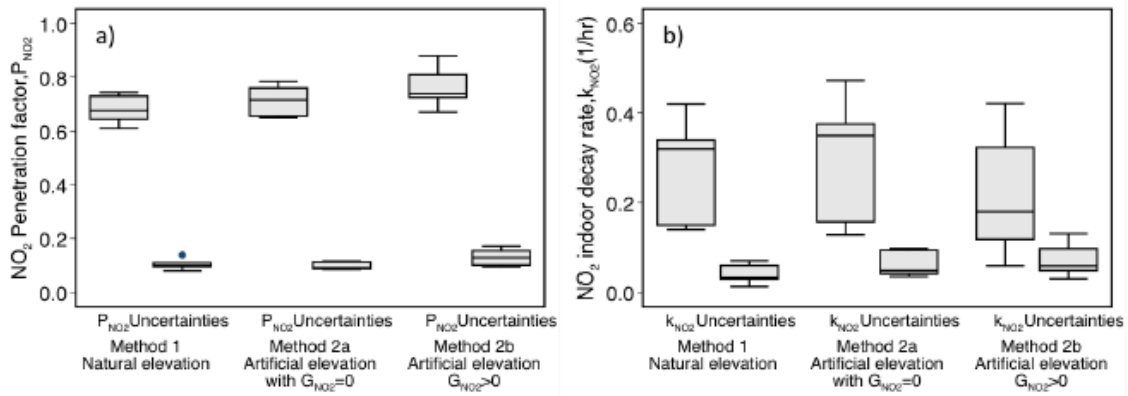


Figure 15. Estimates and absolute uncertainties estimated by three different methods for (a) NO<sub>2</sub> penetration factor and (b) NO<sub>2</sub> indoor loss rate.

### Influence of indoor environmental and ambient conditions

The resulting estimates and uncertainties of the penetration factor ( $P$ ) and loss rate ( $k$ ) for NO<sub>2</sub> and NO<sub>x</sub> from the 12 tests were analyzed in relation to several indoor and outdoor environmental factors. No significant relationships were identified between  $P_{NO_2}$  and any of these variables (Spearman's rank correlations,  $p > 0.05$ ). However,  $k_{NO_2}$  showed a stronger correlation with the indoor humidity ratio ( $R^2 = 0.61$ ) compared to indoor relative humidity (RH) ( $R^2 = 0.53$ ), as illustrated in Figure 16.

Additionally, the relationship between indoor NO loss rates ( $k_{NO}$ ) and ambient ozone concentrations, as well as estimated indoor ozone concentrations during testing, was examined. Figure 17a shows the  $k_{NO}$  estimates from each test plotted against the average outdoor ozone concentration measured at the nearest regulatory monitor. Figure 17b displays the  $k_{NO}$  estimates plotted against the estimated average ozone concentration in the infiltrating air. The latter was calculated by multiplying the average outdoor ozone concentration during testing by the measured air exchange rate (AER) and the average ozone penetration factor ( $P_{O_3} = 0.54$ ) for the test apartment, which was determined in the prior ozone part. This estimate serves as a proxy for the likely indoor ozone concentration during testing, as actual concurrent indoor O<sub>3</sub> measurements were not taken.

Comparing Figures 17a and 17b reveals that indoor NO loss rates ( $k_{NO}$ ) were more strongly correlated with the estimated ozone concentration in the infiltrating air ( $R^2 = 0.95$ ) than with ambient outdoor ozone concentrations ( $R^2 = 0.47$ ). However, the



slopes on both regressions ( $0.05$  and  $0.17 \text{ ppb}^{-1} \text{ h}^{-1}$ , respectively) were lower than the previously reported reaction rate constant between  $\text{NO}$  and  $\text{O}_3$  ( $1.8 \text{ ppb}^{-1} \text{ h}^{-1}$ ). This suggests that indoor  $\text{NO}$  loss was not primarily driven by direct reactions with ozone but likely involved other unmeasured oxidants, such as hydroperoxyl and alkylperoxy radicals, as well as the hydroxyl radical.

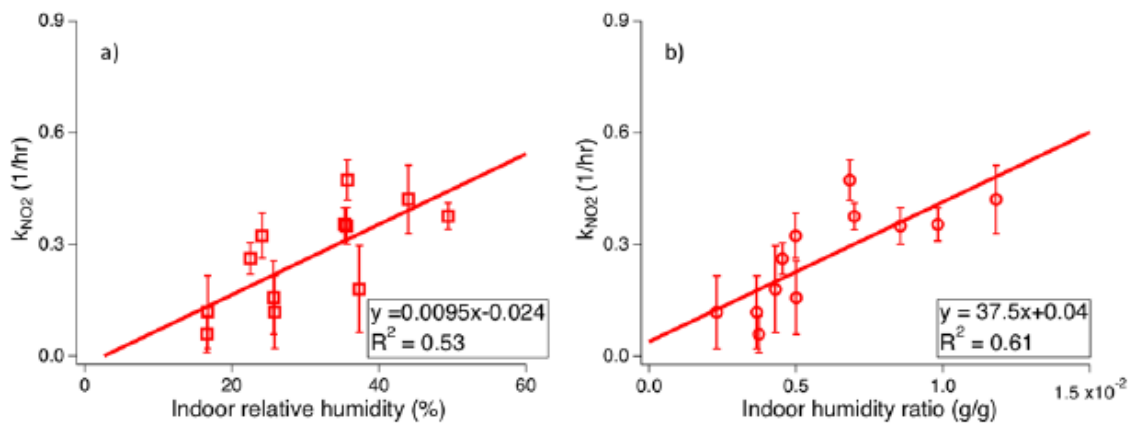


Figure 16.- Estimates of indoor  $\text{NO}_2$  loss rates versus (a) average indoor relative humidity and (b) average indoor humidity ratio during testing.

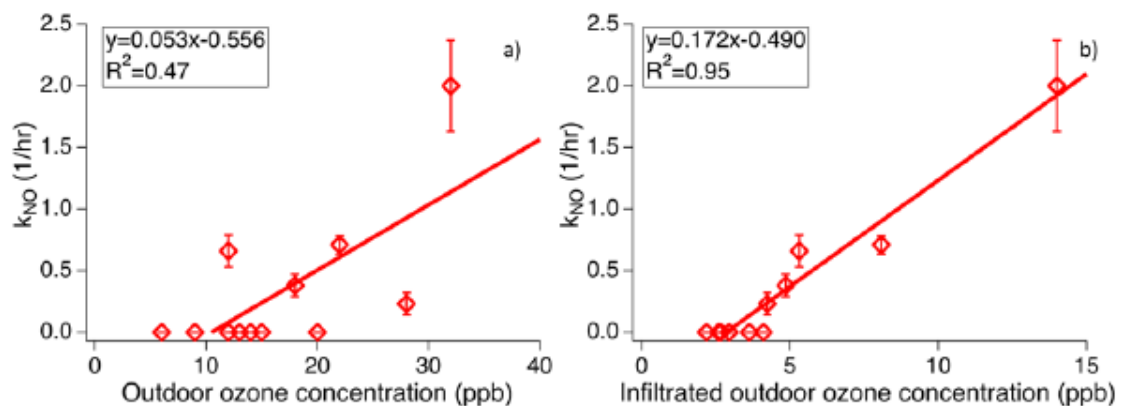


Figure 17.- Estimated indoor  $\text{NO}$  loss rates versus (a) average outdoor ozone concentrations measured at the nearest regulatory monitor  $\sim 10 \text{ km}$  away and (b) estimated average concentration of infiltrated outdoor ozone.

#### 5.4.- Field measurements of outdoor pollutant penetration in a diverse sample of residential buildings in Chicago, IL

The test methods developed and refined in objectives 1 through 3 were first evaluated for their practicality in field measurements. The methods described beforehand in 5.3.- Novel methods to measure the penetration factor of nitrogen oxides for measuring  $\text{NO}_x$  penetration factors require long-term indoor and outdoor

monitoring in unoccupied residences, making them less feasible for immediate field use. However, the methods refined in 5.1.- A refined method to measure the ozone penetration factor in residences under infiltration conditions and 5.2.- An improved method to rapidly measure the penetration of fine and ultrafine particles in unoccupied residences for measuring the penetration factors of ozone, as well as fine and ultrafine particles, generally required only a 4 – 5 hour testing period in unoccupied residences. These shorter tests can be combined and applied more broadly in field measurements. Consequently, these methods were used in a diverse sample of 22 homes in Chicago, IL. This sample included 9 single-family residences and 13 multi-family units of varying ages and envelope materials. Additionally, given the growing implementation of energy efficiency retrofits, such as air sealing and insulation, which alter air infiltration pathways, a subset of the field measurements was conducted in 13 homes before and after these retrofits. This subset included 8 single-family homes and 5 multi-family units or apartments. The following tables provide a full list of the homes tested, both with and without retrofits.

*Table 1.- Summary of test homes with retrofits.*

HOME #	CITY	AREA [ft <sup>2</sup> ]	YEAR BUILT	SF / MF	VISIT 1 (pre)	VISIT 2 (post)	RETROFITS
1	Des Plaines	1620	1951	SF	08/26/2015	09/09/2015	Attic air sealing + attic insulation (blown in cellulose)
2	Lombard	1750	1932	SF	09/23/2015	11/02/2015	Attic air sealing and insulation up to R-49
3	Park Ridge	1821	1956	SF	11/09/2015	11/16/2015	Attic air sealing and insulation
4	Evanston	1441	1924	SF	02/22/2016	04/22/2016	Attic air sealing and insulation
5	Chicago	870	1894 (renovated)	MF 2 flat	02/25/2016	03/07/2016	Attic air sealing and insulation
6	Chicago	780	1894 (renovated)	MF 2 flat	02/26/2016	n/a	Attic air sealing and insulation
7	Chicago	1000	1894 (renovated)	MF 2 flat	02/26/2016	03/07/2016	Attic insulation + air sealing in upstairs unit

8	Chicago	1078	1915	SF	03/02/2016	04/13/2016	Attic rigid and blown in insulation + blown in wall insulation into balloon framing wall cavities + some crawlspace rigid insulation
9	Chicago	876	1910	MF 2 flat	07/27/2016	08/23/2016	Attic air sealing and insulation and weather stripping on doors
10	Chicago	876	1910	MF 2 flat	07/26/2016	08/22/2016	Attic air sealing and insulation and weather stripping on doors
11	Chicago	1845	1959	SF	08/19/2016	10/14/2016	Closed cell foam air sealing attic floor; blown in cellulose to R-49 (up to 15 inches), insulate attic hatch, air seal furnace room
12	Chicago	2448	1921	SF	01/23/2017	03/03/2017	New insulated windows replacement only
13	Chicago	1224	1901	SF	12/20/2017	02/22/2017	Sprayed in insulation; drilled holes in top and bottom of walls; main floor and upper level exterior side walls; attic insulation

Table 2. - Summary of test homes without retrofits.

HOME #	CITY	AREA [ft <sup>2</sup> ]	YEAR BUILT	SF / MF	VISIT 1 (pre)	VISIT 2 (post)	RETROFITS
S1	Chicago	667	1971	MF - apartment	42649	n/a	None
S2	Chicago	441	1968	MF - apartment	06/21/2016	n/a	None
S3	Chicago	910	1888	MF - apartment	07/19/2016	n/a	None
S4	Chicago	753	1885	MF 2 flat	08/13/2016	n/a	None
S5	Chicago	1358	1915	MF - apartment	10/03/2016	n/a	None
S6	Chicago	441	1968	MF - apartment	08/13/2017	n/a	None
S7	Chicago	1150	1969	MF - apartment	11/01/2017	n/a	None
S8	Chicago	2340	1989	SF	03/08/2018	n/a	None
S9	Chicago	1400	1885	MF duplex	06/06/2018	n/a	None

A combined instrumentation system, incorporating the particle and ozone monitors, was assembled using a medium-sized, roller-equipped audio rack case, which was typically positioned in the living room near a window. The particle instruments were connected using conductive silicon tubing, and they shared the switching system outlined previously. Meanwhile, the ozone monitor was connected to a separate switching system, using PTFE inline tubing.

At each visit, testing began around 8:00 AM. Equipment setup and orientation typically took about an hour. After setup, a blower door test was conducted to depressurize the house, with all exterior windows and doors closed. Three parameters – effective leakage area (ELA), normalized leakage (NL) and air change rate at a pressure difference of 50 pascals (ACH 50) – were calculated to evaluate the airtightness of the building. Following this, the blower door fan was used to pressurize the house with windows and doors open, allowing cross-ventilation to introduce outdoor particles and ozone into the indoor environment. The objective was to achieve approximately 90% of the outdoor concentrations of both PM and ozone. Once the desired pollutant levels were reached, all exterior openings were closed, and the building was vacated by both researchers and residents.

The switching systems were configured to measure indoor particle and ozone concentrations for approximately 40 – 60 minutes to capture the initial indoor decay of both pollutants. Indoor and outdoor particle and ozone concentrations were then measured alternately at high time-resolution intervals (4 minutes indoors and outdoors for PM, and 2 minutes indoors and outdoors for ozone) over a period of about 5 hours. The air exchange rate (AER) during the testing period was measured using the CO<sub>2</sub> elevation decay method, as described in previous sections.

#### 5.4.1.- Example of test data

Figure 18 shows examples of test results for both (a) ozone and (b) ultrafine particle (UFP) elevation and decay during the post-visit on April 13, 2016, at test home 8. The air exchange rate (AER) during this test was  $0.22 \pm 0.01 \text{ h}^{-1}$ . Using the methods outlined, the decay rates ( $k$ ) for ozone and UFP were estimated at  $4.32 \pm 0.07 \text{ h}^{-1}$  and

$0.79 \pm 0.03 \text{ h}^{-1}$ , respectively. These values were then applied to the solution methods to calculate penetration factors, resulting in estimates of  $0.75 \pm 0.14$  for ozone and  $0.68 \pm 0.07$  for UFP.

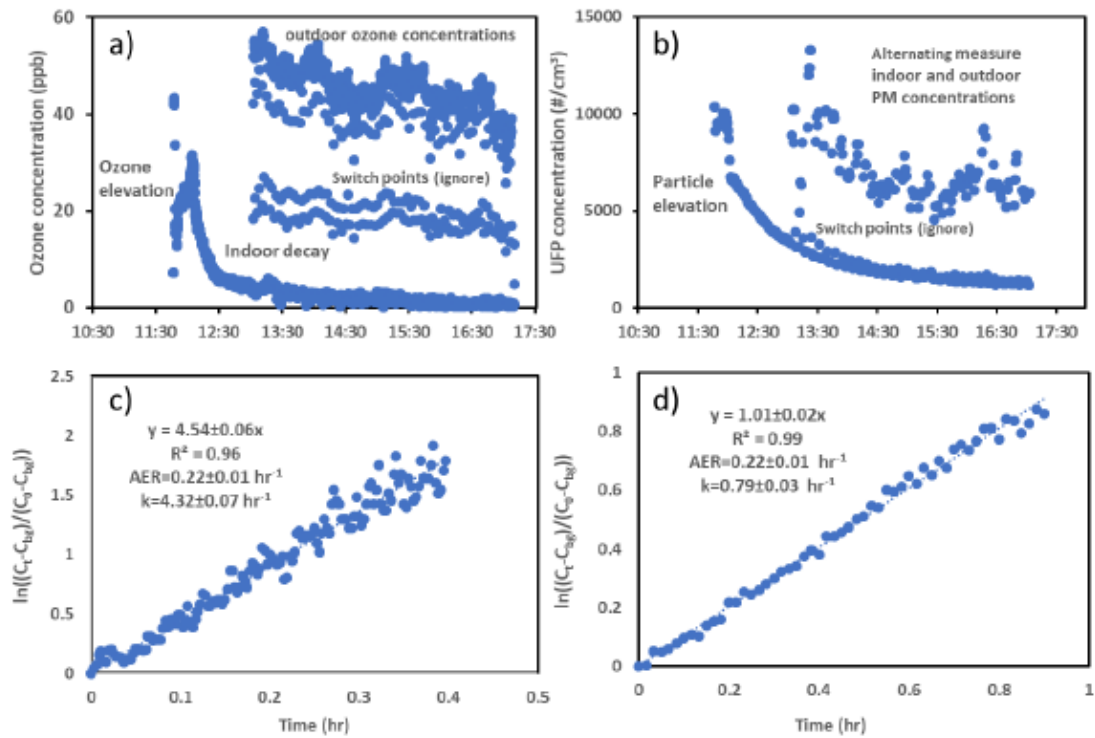


Figure 18.- (a) time-series indoor and outdoor ozone concentrations, (b) time-series UFP number concentration data from the same test period, (c) log-linear regression used to estimate the indoor first order ozone decay rate constant, and (d) log-linear regression used to estimate the indoor UFP deposition loss rate constant.

#### 5.4.2.- Pollutants penetration factors in single family homes and multi-family units

Figure 19 presents the results of measured penetration factors for PM<sub>2.5</sub>, ultrafine particles and ozone in both single-family homes and multi-family units. The average penetration factors for PM<sub>2.5</sub> (mean  $\pm$  SD) in single-family homes, which included 17 successful tests across 9 homes (8 before and after retrofits, and 1 without retrofits), were  $0.80 \pm 0.09$ , with values ranging from 0.65 to 1.02. In multifamily units, the average PM<sub>2.5</sub> penetration factor (from 16 successful tests in 12 homes, 4 tested before and after retrofits and 8 without retrofits) was  $0.90 \pm 0.12$ , with a range of 0.62 to 1.03.

Similarly, for ultrafine particles, the average penetration factor in single-family homes (17 successful tests across 9 homes) was  $0.70 \pm 0.11$ , with a range of 0.53 to 0.87. In multi-family units (16 successful tests in 12 homes), the average penetration factor was  $0.75 \pm 0.16$ , ranging from 0.46 to 0.95.

The average ozone penetration factors were estimated at  $0.73 \pm 0.16$  for single-family homes (13 successful tests across 7 homes) and  $0.71 \pm 0.23$  for multi-family units (7 successful tests in 6 homes). Wilcoxon rank-sum tests indicated no significant differences ( $p < 0.05$ ) between single-family homes and multi-family units for the penetration factors of fine particles, ultrafine particles and ozone. However, fewer ozone penetration tests were successful due to low outdoor concentrations that often fell below the detection limits of the instruments.

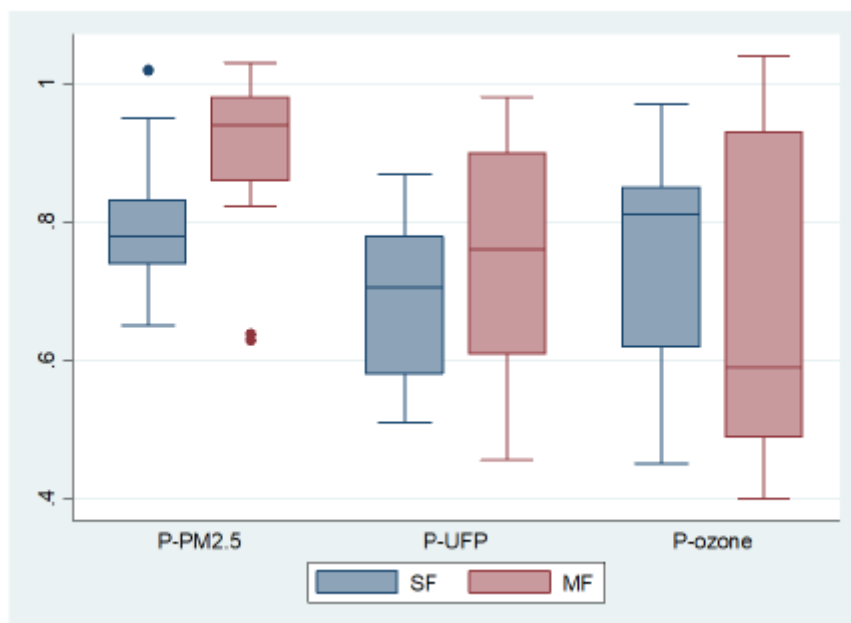


Figure 19.- Penetration factors estimated for mass concentrations of particle less than PM2.5, UFPs and ozone measured in single-family homes and multi-family units in and around Chicago, IL.

### 5.4.3.- Results of energy efficiency retrofits

Figure 20a presents the results of the air exchange rate at 50 pascals pressure difference (ACH50), calculated from blower door data for 12 homes that underwent testing both before and after retrofits. The blower tests indicate that energy efficiency retrofits reduced ACH50 by an average of  $16 \pm 17\%$ , with reductions

ranging from approximately 0% to 46%. As a result, the test homes were classified as moderately tightened by the retrofits.

Figure 20b shows the changes in air exchange rates (AER) during testing before and after the retrofits, with variations ranging from a 39% decrease to a 26% increase, averaging a  $16 \pm 19\%$  reduction. The air exchange rates were influenced by factors such as airtightness, indoor and outdoor temperature differences and wind conditions during the test periods. A moderate correlation ( $R^2 = 0.39$ ) was found between changes in ACH50 and changes in AER, primarily due to variations in these driving forces.

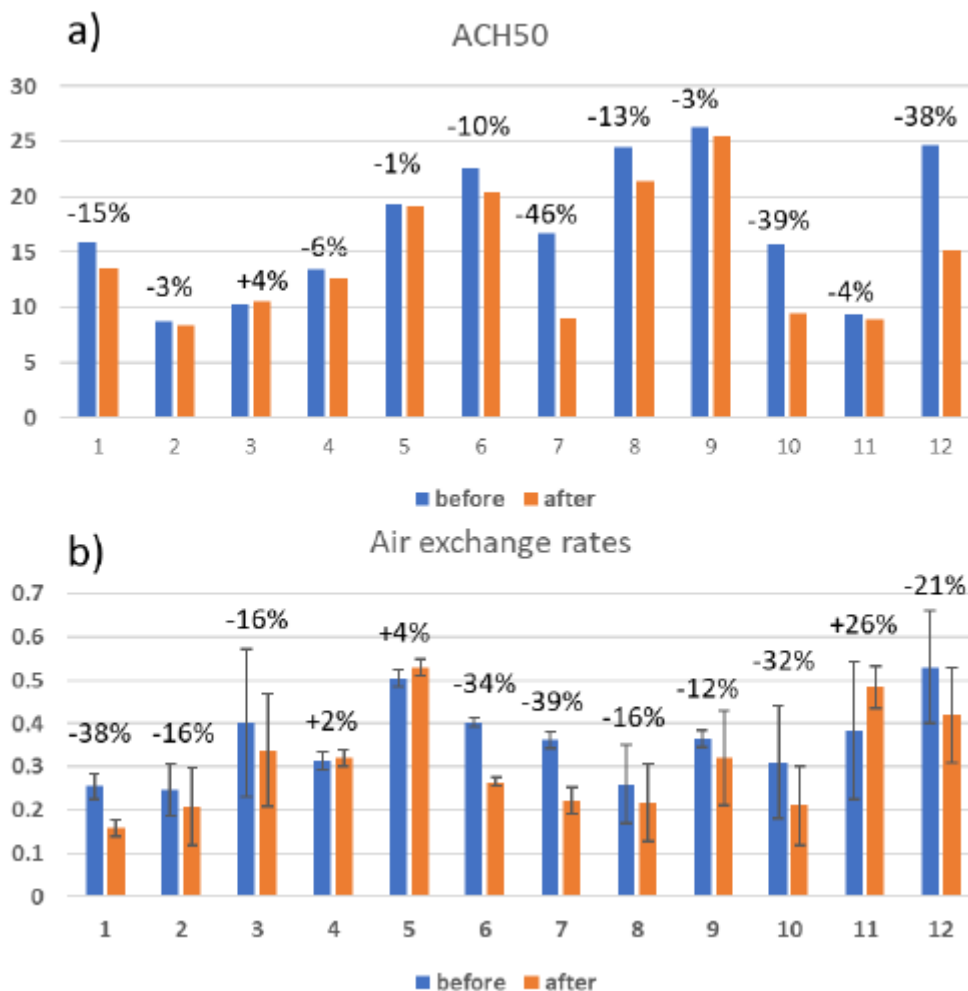


Figure 20.- (a) Air exchange rate at 50 pascal pressure difference (ACH50) and (b) air exchange rates measured during tests for homes before and after retrofits.

#### 5.4.4.- Results of infiltration factors and penetration factors before and after retrofits

Figure 21a illustrates the average ( $\pm$  S.D.) infiltration factors for three pollutants measured before and after retrofits. For PM<sub>2.5</sub>, the average infiltration factor was  $0.40 \pm 0.11$  before the retrofits and  $0.39 \pm 0.12$  afterward. Similarly, the infiltration factor for ultrafine particles (UFPs) remained consistent at  $0.22 \pm 0.04$  both before and after the retrofits. The infiltration factor for ozone was slightly higher, with averages of  $0.05 \pm 0.02$  before and  $0.06 \pm 0.03$  after retrofits.

No significant differences were observed in the infiltration factors before and after retrofits, as determined by the Wilcoxon signed-rank test. This lack of change may be due to the influence of various factors, such as envelope tightness, weather conditions during testing and pollutant characteristics, including particle size distributions and ozone reaction rates.

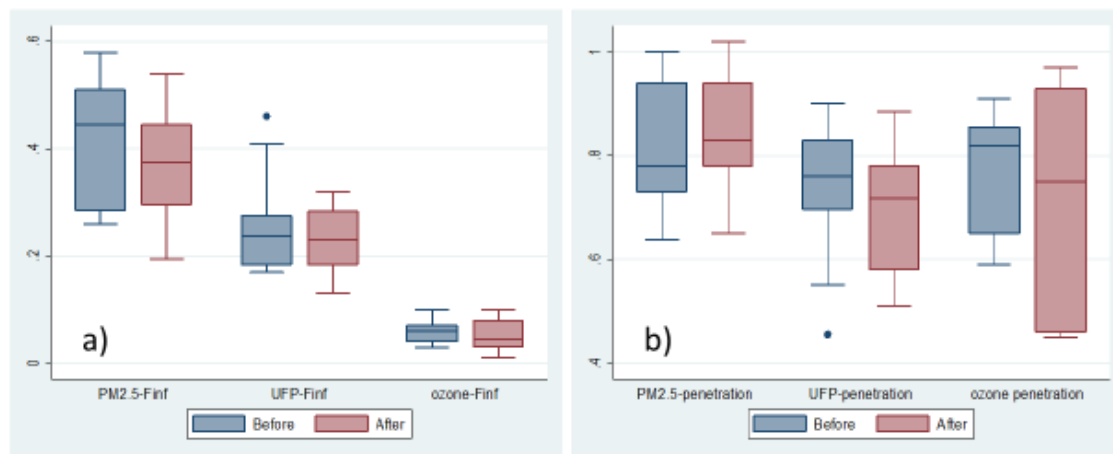


Figure 21.- (a) Infiltration factors and (b) penetration factors estimated for mass concentrations of particle less than PM<sub>2.5</sub>, UFPs and ozone measured in test homes before and after energy efficiency retrofits applied.

Figure 21b presents the estimated penetration factors for three pollutants before and after retrofits. The average ( $\pm$  S.D.) penetration factor for PM<sub>2.5</sub> was  $0.83 \pm 0.11$  prior to the retrofits and  $0.85 \pm 0.10$  afterward. For ultrafine particles (UFPs), the average penetration factor decreased slightly from  $0.74 \pm 0.13$  to  $0.70 \pm 0.11$  following the retrofits. Similarly, for ozone, the penetration factor dropped from  $0.76 \pm 0.11$  before the retrofits to  $0.70 \pm 0.20$  afterward.





No significant differences in penetration factors were observed before and after the retrofits, as determined by the Wilcoxon signed-rank test. Weak correlations were found between changes in the penetration factors for the three pollutants and changes in envelope airtightness. The R-squared values for correlations between changes in effective leakage area (ELA) and the penetration factors for UFPs, PM<sub>2.5</sub> and ozone were 0.31, 0.19 and 0.11, respectively. This suggests that the moderate envelope tightening of around 16% from the retrofits did not significantly alter the ability of pollutants to penetrate through the building envelope.

To further investigate the impact of indoor and outdoor conditions on pollutant penetration, Figure 22 shows plots of changes in penetration factors for (a) PM<sub>2.5</sub> and (b) UFPs against changes in indoor-outdoor temperature differences. Negative correlations with R-squared values of 0.40 and 0.56 suggest that a decrease in temperature difference between indoors and outdoors likely increases the penetration factors for both fine and ultrafine particles in the same dwellings. This can be explained by the fact that during testing, indoor temperatures were generally higher than outdoor temperatures. A larger temperature difference leads to a stronger stack effect in the home, influencing pollutant infiltration pathways, often through the basement or crawlspace, which reduces the infiltration of outdoor pollutants. Since the retrofits mostly involved attic air sealing and insulation but not basement or crawlspace sealing, the primary infiltration pathways likely remained unaffected by the retrofits, limiting the impact on pollutant penetration.

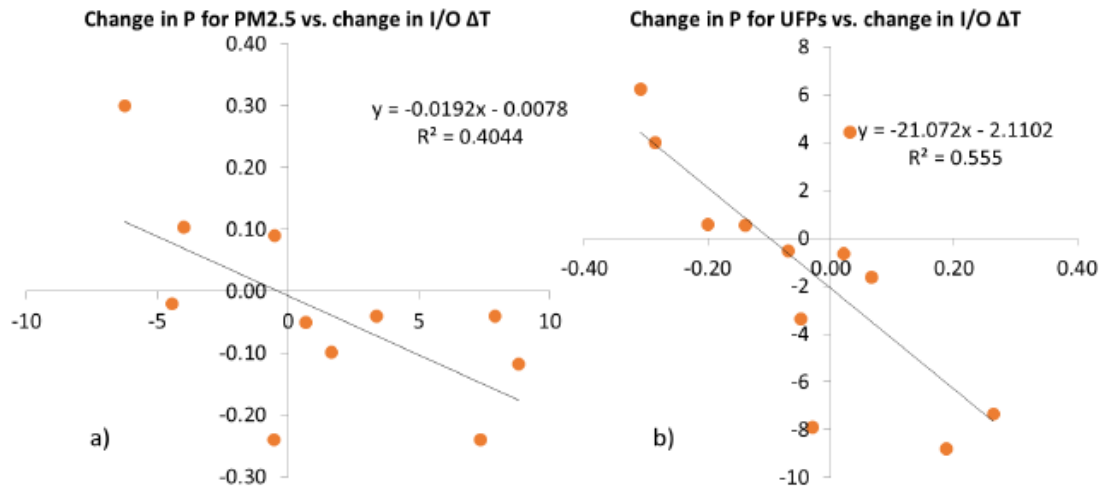


Figure 22.- Plots of changes in penetration factors for (a) PM2.5 and (b) UFPs versus changes in differences of indoors and outdoors temperature during the tests in homes before and after retrofits.

## 6.- PRACTICAL IMPLICATIONS

The findings of this dissertation offer valuable insights into the infiltration and penetration of outdoor pollutants into residential buildings, with implications for both building design and public health. The refined methodologies and new data can serve as tools for improving indoor air quality (IAQ) assessments, informing policies and guiding construction practices. This section discusses the practical applications of the research findings and outlines possible future directions for further exploration.

### 6.1.- Application in building design and construction

One of the most significant practical implications of this work is the potential to influence building design and construction practices. The research demonstrates how different building envelope characteristics, such as materials and airtightness, can significantly affect the penetration of outdoor pollutants like ozone, particulate matter (PM<sub>2.5</sub>) and nitrogen oxides (NO<sub>x</sub>). Architects, engineers and builders can use these findings to design more airtight and energy-efficient buildings while also mitigating pollutant infiltration.

For instance, the results suggest that improving airtightness alone is not sufficient to reduce pollutant penetration in all cases, as seen in the limited impact of energy-efficient retrofits. This highlights the importance of combining airtightness measures with proper ventilation strategies, such as mechanical ventilation with high-efficiency particulate air (HEPA) filters, to reduce pollutant exposure in indoor certifications, such as LEED or WELL, and inform local building codes focused on health and energy efficiency.

### 6.2.- Policy and regulatory implications

The data generated by this research can also be valuable in shaping policies that aim to improve public health through better IAQ. Regulatory bodies could adopt the refined test methods as standardized protocols for evaluating pollutant infiltration in residential and commercial buildings. These standards could become a key

component of IAQ guidelines, ensuring that homes, particularly in urban areas with high levels of outdoor pollution, meet minimum requirements for protecting occupants from harmful airborne substances.

Additionally, the findings could support initiatives aimed at reducing health disparities. Low-income housing, which often lacks the resources for energy-efficient buildings in areas with high outdoor pollution levels, ensuring that vulnerable populations benefit from improved indoor environments.

### 6.3.- Public health applications

This research has the potential to inform public health strategies aimed at reducing the overall exposure of individuals to outdoor pollutants. Since much of human exposure to outdoor-origin pollutants occurs indoors, understanding the dynamics of pollutant infiltration is crucial for accurately estimating population exposure levels. By incorporating building-specific infiltration data, health risk models could be refined to provide more accurate predictions of long-term health impacts, such as respiratory and cardiovascular diseases associated with PM<sub>2.5</sub> and ozone exposure.

Public health authorities could also use these findings to develop targeted recommendations for building occupants during periods of high outdoor pollution, such as during wildfires or high ozone days. For example, individuals living in homes with higher infiltration rates could be advised to use air purifiers or avoid opening windows when outdoor pollution levels are elevated.



## 7.- FUTURE RESEARCH DIRECTIONS

While this dissertation provides a robust framework for understanding the infiltration of outdoor pollutants into residential environments, several avenues remain unexplored and present opportunities for further research. Expanding on these findings could significantly enhance our understanding of indoor air quality and inform both policy and practical interventions aimed at mitigating health risks associated with poor IAQ. Future research could explore the following areas.

### 7.1.- Impact of climate variability

Climate conditions such as temperature, humidity and seasonal shifts can have a substantial influence on pollutant penetration and indoor air quality. In regions increasingly affected by climate change with rising temperatures and more frequent extreme weather events, the dynamics of pollutant infiltration may shift. Further studies could focus on the impact of varying climate conditions on penetration factors, testing how fluctuations in outdoor environments alter infiltration patterns. This research would be particularly relevant for regions undergoing rapid environmental changes, where building designs may not yet be adapted to the new realities posed by climate extremes.

### 7.2.- Longitudinal studies on pollutant penetration

While the current research provides valuable snapshots of pollutant infiltration, long-term studies are necessary to observe these patterns evolve over time. By conducting longitudinal studies, researchers could monitor how infiltration rates change due to environmental factors, building wear and tear, or technological advancements like energy-efficient retrofits. Longitudinal data would offer insights into the durability and sustained effectiveness of various retrofitting strategies, as well as help track how chronic exposure to pollutants influences indoor air quality over years or decades.

### 7.3.- Expanding the scope to commercial and public buildings

Although this study focuses on residential buildings, a significant portion of the population spends considerable time in commercial buildings, educational institutions and healthcare facilities. These environments differ from homes in terms of building design, usage patterns and ventilation systems. Future research should aim to apply similar methodologies to these types of structures, particularly schools and hospitals, where vulnerable populations such as children, the elderly and immuno-compromised individuals may face higher health risks from exposure to outdoor-origin pollutants.

### 7.4.- Health outcome correlations

While pollutant infiltration measurements provide crucial data on indoor air quality, future research could deepen our understanding by correlating these measurements with health outcomes in building occupants. Epidemiological studies could be conducted to explore the relationships between long-term exposure to infiltrated pollutants and health issues such as respiratory diseases, cardiovascular conditions, or cognitive impairments. Special attention could be given to vulnerable populations – children, the elderly and those with pre-existing health conditions. By establishing these correlations, future studies could provide strong evidence for policy changes aimed at improving air quality standards in buildings.

### 7.5.- Evaluation of advanced filtration technologies

As new air filtration and purification technologies emerge, it becomes important to evaluate their effectiveness in reducing indoor concentrations of outdoor pollutants. Future studies could test various mechanical ventilation systems equipped with HEPA filters, as well as cutting-edge technologies like UV germicidal irradiation or photocatalytic oxidation. Understanding how these technologies perform in real-world settings, particularly in homes or buildings located in highly polluted areas, would provide valuable insights for improving air quality in both residential and commercial settings.

## 8.- CONCLUSIONS

This dissertation presents a combination of refined and new testing methods aimed at assessing the infiltration and penetration of outdoor pollutants into residential indoor environments. These methods were applied in both a test apartment unit and a variety of single-family homes and multi-family units, each exhibiting diverse building characteristics. The research involved four key investigations, each aligned with specific research objectives.

Investigation 1 refined a test methodology from previous studies by incorporating the latest instrumentation to measure the penetration of ambient ozone into residential buildings under infiltration conditions. This method was applied in a test apartment unit. Investigation 2 further refined and implemented a methodology for rapidly measuring the size-resolved penetration of ambient fine and ultrafine particulate matter into buildings, also using the test apartment unit. Investigation 3 developed and applied novel techniques for measuring the penetration factor of nitrogen oxides into the test apartment, based on long-term indoor and outdoor measurements. This study also explored the effects of indoor and outdoor environmental conditions on nitrogen oxide penetration and indoor loss. Finally, investigation 4 combined the refined methods for Objectives 1 and 2 and deployed an integrated instrumentation system to conduct field tests measuring infiltration and penetration factors for ozone and fine and ultrafine particles in a range of residential buildings in and around Chicago, IL.

The key contributions and findings from the four investigations in this dissertation are summarized below:

1. A refined method for measuring the penetration factor of ozone was successfully developed and applied in an unoccupied test apartment unit. This study represents the first known instance of measuring ozone penetration factors through building envelopes under normal infiltration conditions. The results showed that the ozone penetration factor in the



- apartment unit was lower than the commonly assumed value of unity ( $P = 1$ ), with an average of  $0.54 \pm 0.10$  across various conditions.
2. A method for more rapid and accurate measurement of size-resolved penetration factors for fine and ultrafine particles was refined and applied in the same test apartment unit. This was also the first known study to experimentally measure the penetration factor for integrated PM<sub>2.5</sub> using time-resolved instrumentation. The method was designed to reduce the testing duration to around five hours without compromising accuracy, offering a practical solution for broader application in field measurements across larger variety of buildings.
  3. Novel methods were developed and applied to measure the penetration factor and indoor loss rates of ambient NO<sub>x</sub> constituents using time-resolved measurements in an unoccupied apartment unit. This is the first known study to quantify nitrogen oxide penetration through building envelopes. The average ( $\pm$  S.D.) NO<sub>2</sub> penetration factor from repeated tests was  $0.72 \pm 0.06$ , which is lower than the commonly assumed value in previous studies. Additionally, the investigation found that NO<sub>2</sub> indoor loss rates were strongly correlated with the indoor humidity ratio, while NO loss rates were closely linked to the estimated ozone concentration in the infiltrating air.
  4. The methods for measuring the penetration factors of ozone, fine and ultrafine particles were further applied to field tests in a variety of residential buildings, including 13 single-family homes and 9 multi-family units, featuring a wide range of construction types and materials. A subset of these homes was tested before and after energy-efficient retrofits to evaluate the impact on pollutant infiltration and penetration. Results showed that the average penetration factors for PM<sub>2.5</sub>, ultrafine particles and ozone in single-family homes were  $0.80 \pm 0.09$ ,  $0.70 \pm 0.11$  and  $0.73 \pm 0.16$ , respectively, while for multi-family units, they were  $0.90 \pm 0.12$ ,  $0.75 \pm 0.16$  and  $0.71 \pm 0.23$ , respectively. The energy-efficient retrofits, which moderately tightened the building envelopes, did not significantly influence the infiltration or penetration of pollutants in this sample of homes.



In conclusion, the research presented in this dissertation offers new test methods and valuable data for evaluating the infiltration of outdoor pollutants into residential buildings. These novel methods can be standardized and adapted for broader application across a wider range of buildings. A key goal of the work was to develop techniques that allow for faster, more efficient and cost-effective assessments of indoor air quality (IAQ). The findings from the field tests can contribute to improving population exposure models by providing more accurate estimates of indoor concentrations of outdoor pollutants. Continued research in these areas is necessary to advance these objectives further.

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