CHARACTERIZING AND MODELING CLOSE-PROXIMITY EXPOSURE TO AN
AIR POLLUTION SOURCE IN NATURALLY VENTILATED RESIDENCES

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ABSTRACT

Near an active indoor emission source, air pollutant levels are elevated and highly-variable, due to non-instantaneous mixing – this causes great uncertainty in estimating a person’s exposure level. This research investigated the magnitude and variability of short-term exposures close to an active point source inside 2 homes, under a range of natural ventilation conditions.

The findings from a newly-developed monitor signal reconstruction method were applied to measurements from 30-37 real-time monitors to capture the spatial and temporal variations of concentrations over 30-min CO tracer gas releases. For 11 experiments involving 2 houses, with natural ventilation conditions ranging from <0.2 to >5 air changes per h, an eddy diffusion model was used to determine the turbulent diffusion coefficients, which ranged from 0.001-0.015 m²s⁻¹. The air change rate showed a significant positive linear correlation ($R^2=0.94$) with the air mixing rate, defined as the turbulent diffusion coefficient divided by a squared length scale representing the room size.

To predict the magnitude of exposure close to an active source, an indoor dispersion model was formulated, invoking the theory of random walk, and incorporating the physical processes of anisotropic turbulent diffusion, removal of the air pollutant, and air pollutant wall reflection. Then, to capture the variability of concentrations in close proximity to an active source, a new piece-wise random walk algorithm was developed to stochastically simulate the transient directionality of emitted plume. The distribution of different exposure cases generated using this model reasonably covered the range of experimental measurements collected in 2 houses, while preserving ensemble averages satisfying the principle of Fickian diffusion.
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CHAPTER 1

INTRODUCTION

1.1 MOTIVATION

People spend 65-70% of the time indoors at home, so personal exposure to residential indoor air pollution constitutes a significant fraction of total exposure (Leech et al., 2002; Briggs et al., 2003; Brasche and Bischof, 2005). Exposure to in-home emission sources has been modeled by the well-mixed mass balance model (e.g. Burke et al., 2001), assuming uniform concentration in space. Since the transient imperfect mixing period immediately after a release of pollutant is typically less than 1 h (Baughman et al., 1994; Drescher et al., 1995; Klepeis, 1999), this model can provide simple and accurate exposure estimates when the source emission and mixing time scales are much smaller than the duration over which the time-averaged concentration (the estimate of exposure) is considered. However, for a continuous source releasing air pollutants over a duration comparable to the exposure time of interest, the imperfect mixing during the emission period becomes important to consider. During this active source period, exposures in close proximity to the source are expected to be substantially higher than those further away from it – this source “proximity effect” cannot be captured by the uniform mixing model commonly used in residential indoor exposure studies.

To examine the proximity effect, controlled experiments using multiple real-time monitors have been conducted to capture the spatial and temporal variation of air pollutant concentrations in residences. They show that exposures within 2 m from the source were up to ~4 times as high as the predictions of the well-mixed mass balance model (Furtaw et al., 1996; McBride et al., 1999, 2002). Most recently, Acevedo-Bolton et al. (2010) reported even higher elevations in concentration close to the source, and pronounced fluctuations with time due to transient directional air movements of turbulent mixing indoors. These results imply that exposures in close proximity to the source are elevated and highly variable.
To model the proximity effect that has been observed in residential environments, one can apply analytical models involving isotropic turbulent diffusion, found in the occupational literature (Wadden et al., 1989; Conroy et al., 1995; Drivas et al., 1996; Demou et al., 2009), to predict the levels of higher exposure close to a household emission source. These models assume that for indoor spaces enclosed by walls, there is no pronounced and persistent directional advection. Air pollutant transport is mainly driven by turbulent eddy motions in the air. These random motions of air allow air pollutants to be dispersed symmetrically with respect to the source, with magnitudes of mixing 100-10000 times as large as the molecular diffusion (Keil, 2000).

The production of the turbulent eddies is related to the inputs of kinetic energy such as air flow coming from windows and doors, and/or from the operating HVAC and fans (Drescher et al., 1995). It is also associated with the inputs of thermal energy such as sunlight heating on wall surfaces and operating space heaters/stoves, responsible for the buoyancy-driven air motions (Baughman et al., 1994). Initially, large eddies (i.e. comparable to the room dimensions) are formed in close proximity to the energy sources introducing transient directional air flow. As these eddies cascade with time from the initial largest sizes to length scales smaller than the size of the plume (Thatcher et al., 2004), the directionality of air pollutant transport becomes less and less noticeable in the room, and can be represented as a turbulent mixing process.

In these models, isotropic turbulent diffusion coefficients \( K \) are used to characterize the magnitudes of the eddy/turbulent diffusion indoors and to represent how fast the spatial spread of the air pollutant grows with time (Fischer et al., 1979). These parameters are typically determined by field experiments (Scheff et al., 1992; Conroy et al., 1995; Demou et al, 2009) and have been found to be positively associated with the amount of kinetic (Drescher et al., 1995) and thermal (Baughman et al, 1994) energy inputs, but limited by the vertical temperature stratification indoors (Drivas et al., 1996).

However, the parameter \( K \) used in these models has not been previously assessed for residential settings to predict higher exposures close to an in-home source. Furthermore,
it never has been possible to model the observed great variability of exposure close to the source using these deterministic approaches.

The main purpose of the thesis is to characterize and model the close-proximity exposure to an active air pollution point source in naturally ventilated residential indoor environments. Using our monitoring array along with a monitor signal reconstruction method, my first goal is to experimentally deduce accurate estimates of turbulent diffusion coefficients in residences under a range of natural ventilation conditions. This characterization allows the use of the existing indoor eddy diffusion models for residential exposure applications. Building on the indoor turbulent diffusion formulation, my second goal is to develop a stochastic exposure model that can describe not only the elevations but also the high variability in close-proximity exposures to an active indoor air pollution source.

1.2 DISSERTATION OVERVIEW

This dissertation is comprised of 5 chapters and 2 appendices. Chapter 1 presents the motivation and overview of the dissertation. Chapter 2 investigates the robustness of my monitor signal reconstruction method for providing accurate close-to-source measurements for the subsequent indoor monitor array experiments. Chapter 3 utilizes the indoor monitor array to characterize turbulent diffusion coefficients under different natural ventilation conditions. Chapter 4 and Appendix A investigate the ability of the new stochastic indoor exposure model to predict elevated and highly variable exposures close to the source. Chapter 5 summarizes the major findings from Chapters 2-4.

The following subsections (section 1.2.1-1.2.3) present brief overviews of the 3 major parts of the thesis (Chapter 2-4) aiming to characterize and model the proximity effect in residential indoor environments, describing how these major parts are interconnected.
1.2.1 Reconstructing Accurate Measurements Close to a Source (Chapter 2)
The real-time CO sensors used in our indoor tracer study are compact, passive air
samplers well-suited for collecting indoor measurements without disturbing the air flow.
However, they cannot respond to the changes in environmental concentrations
instantaneously. To obtain accurate measurements of rapidly-fluctuating concentrations
close to the source, I developed a mathematical model that can reconstruct accurate
transient concentration time series from monitor readings. Using this model,
measurement errors associated with different averaging times were quantified. This
already-published study, entitled “Model-based reconstruction of the time response of
electrochemical air pollutant monitors to rapidly varying concentrations,” is presented in
Chapter 2.

Chapter 2 served as the quality assurance step for the indoor tracer study (Chapter 3):
Chapter 2’s monitor calibration and time response testing enabled us to decide on which
and how many monitors to use for the following monitor array experiments presented in
Chapter 3. Chapter 2 also examined the expected concentration fluctuations and the
corresponding monitoring errors for the experimental setup used in Chapter 3, using the
developed signal reconstruction method. This provided direct indications regarding what
averaging time to use to reasonably capture accurate concentration fields in the
subsequent indoor tracer study (Chapter 3).

1.2.2 Experimentally Characterizing the Proximity Effect (Chapter 3)
In indoor models of exposure close to a source, an empirically-adjusted isotropic
turbulent diffusion coefficient is used to capture the magnitude of turbulent mixing in an
indoor space, and how exposures vary with the distance from an active source—that is,
the proximity effect. I used the results from 11 indoor experiments, each using 30-37
monitor to measure a series of controlled CO point source releases, to estimate turbulent
diffusion coefficients ($K$) under different natural ventilation conditions. In addition, I
examined whether $K$ can be predicted using 2 readily-measured parameters: the air
change rate and room dimensions. This study, presented in Chapter 3, has been submitted
for publication as a paper entitled “Modeling exposure close to air pollution sources in
The findings in Chapter 3 offered experimental insights into what assumptions and simplifications can be made for the subsequent development of the indoor exposure/dispersion model (Chapter 4). It also provided a method to predict the turbulent diffusion coefficient ($K$) needed as input for the use of the indoor dispersion model to predict the proximity effect. The temporal and spatial measurements collected in Chapter 3 were subsequently utilized to test how well the model can predict the variability of exposure in the presence of an indoor active emission source.

### 1.2.3 Modeling the Proximity Effect (Chapter 4)

To model the elevated and highly variable exposures in close proximity to an active indoor point source, I developed an indoor exposure model, invoking the random-walk particle tracking method. In this study, I formulated a new piece-wise random walk algorithm to stochastically simulate transient directional air movements of turbulent mixing indoors, responsible for the great variability of exposure close to the source. Simulation results of the new algorithm were compared with the real indoor measurements from Chapter 3. This modeling effort, presented in Chapter 4, is entitled “Modeling the effect of proximity on exposure to an indoor active air pollution source in naturally ventilated rooms: An application of the stochastic random walk process.” The MATLAB script of the random-walk indoor exposure model is provided in Appendix A.

Finally, Chapter 5 summarizes the major findings of this thesis research and recommendations for future investigations.

In addition to the main thesis focusing on the proximity effect, Appendix B includes another exposure-oriented indoor study completed and published during my Ph.D. studies, examining indoor particle resuspension due to foot traffic.
REFERENCES


CHAPTER 2

Model-Based Reconstruction of the Time Response of Electrochemical Air Pollutant Monitors to Rapidly Varying Concentrations\(^1\)


ABSTRACT

Electrochemical sensors are commonly used to measure concentrations of gaseous air pollutants in real time, especially for personal exposure investigations. The monitors are small, portable, and have suitable response times for estimating time-averaged concentrations. However, for transient exposures to air pollutants lasting only seconds to minutes, a non-instantaneous time response can cause measured values to diverge from actual input concentrations, especially when the pollutant fluctuations are pronounced and rapid. Using 38 Langan carbon monoxide (CO) monitors, which can be set to log data every 2 s, we found electrochemical sensor response times of 30-50 s. We derived a simple model based on Fick’s Law to reconstruct a close to accurate time series from logged data. Starting with experimentally measured data for repetitive step input signals of alternating high and low CO concentrations, we were able to reconstruct a much improved 2-s concentration time series using the model. We also utilized the model to examine errors in monitor measurements for different averaging times. By selecting the averaging time based on the response time of the monitor, the error between actual and measured pollutant levels can be minimized. The methodology presented in this study is useful when aiming to accurately determine a time series of rapidly time-varying concentrations, such as for locations close to an active point source or near moving traffic.

\(^1\) A version of this chapter has been published in *Journal of Environmental Monitoring*, 2010, 12, 846-853. Reproduced by permission of The Royal Society of Chemistry http://pubs.rsc.org/en/Content/ArticleLanding/2010/EM/B921806H
2.1 INTRODUCTION

With the development of electronic air monitoring devices, measuring pollutant levels in real time has become feasible for a variety of purposes, ranging from personal exposure measurements to homeland security. Electrochemical monitors, called *amperometric gas sensors*, are one type of instrument commonly used for monitoring the time series of gaseous pollutant concentrations. Typically, these monitors are small, rugged, and have suitable accuracy and precision for industrial settings, personal monitoring, and indoor and outdoor measurement surveys. Hence, they have been used in many non-occupational and occupational settings.

For example, electrochemical gas monitors have been used to assess carbon monoxide (CO) concentrations from motor vehicles (Ott et al., 1994; Gómez-Perales et al., 2004; Zagury et al., 2000; Duci et al., 2003; C.C. Chan et al., 1991; L.Y. Chan et al., 1999, 2001, 2002; A.T. Chan et al., 2003) and environmental tobacco smoke (Ott et al., 1992; Klepeis et al., 1996, 1999; Wallace., 2000; Ott et al., 2008). They have been used to measure nitrogen monoxide, nitrogen dioxide, and/or sulfur dioxide levels emitted during fuel combustion (Berry and Colls, 1990; Kwong et al, 2007), and ammonia emissions from cleaning products (Fedoruk et al, 2005) and livestock (Sommer et al., 2009). A variety of monitors are commercially available from different manufacturers (*e.g.*, Langan Products, Inc., San Francisco, CA, USA; Draeger Safety, Inc., Pittsburgh, PA, USA; Solomat, Ltd, Bishops Stortford, UK; TSI, Inc., Shoreview, MN, USA; Interscan, Inc., Chatsworth, CA, USA; Testo, Inc., Lenzkirch, Germany).

Some electrochemical monitors include internal pumps to draw in air samples actively, while others allow the pollutant to move into the monitor by diffusion. All monitors, regardless of air sampling method, use a built-in electrochemical sensing cell. The sensing cell consists of a working electrode where targeted gases are either oxidized or reduced to generate an electrical current. The resulting current, which is proportional to the gas concentration, is amplified by the external circuit and then transmitted to a built-in data logger and stored (Warburton et al, 1998).
To more accurately reflect the levels of air pollutants, these electrochemical sensing cells are usually coupled to a diffusion-controlled design to ensure complete oxidation/reduction reactions at the electrode (Warburton et al., 1998) as well as to remove unwanted interfering gases (Ott et al., 1986). Air samples, either actively or passively delivered to the sensing cell surface, must diffuse through multiple layers of membranes to reach the interior of the sensing cell, allowing the subsequent electrochemical reactions to take place (Warburton et al., 1998).

For electrochemical monitors, there is a delay between a change in environmental concentration and the sensor’s response. Manufacturers characterize this instrument delay with “$t_{50}$” or “$t_{90}$”, the time required for a sensor to reach 50% or 90%, respectively, of its equilibrium value. For electrochemical CO sensors, $t_{90}$ is typically 20-60 s (Esber et al., 2007). Studies have shown that this delay in monitor response generally follows a first-order behavior (Bay et al., 1974; Sedlak et al., 1976; Esber et al., 2007), so the response time can also be represented by a characteristic time constant, $\tau$, where $t_{50} = 0.693 \tau$ and $t_{90} = 2.3 \tau$. Warburton et al. (1998) analyzed the design and time response of the sensor cell and report that the $t_{50}$ response time is attributable to the diffusion-limited design of the sensing cell, whereas the $t_{90}$ response time depends on both diffusion and the electrical properties of the sensor.

Instruments used for protecting health in the workplace must have sufficiently rapid response times to give early warning of a dangerous buildup of toxic gases. Breath analysis of pollutants also requires short response times. Measurements taken in close proximity to an active source, such as a cigarette, show rapid variations with time – extremely high concentration peaks lasting for only a few seconds, called microplumes (Furtaw et al., 1996; McBride et al., 1999; McBride, 2002; Klepeis et al., 2007, Klepeis et al., 2009). When the fluctuations of pollutant concentrations are pronounced and rapid, the discrepancy between the true input signal and the monitor output signal can be significant. In this case, it is important to determine the accuracy of time-averaged values
computed from the continuous readings, by accounting for the time response of the monitor.

To deal with monitoring inaccuracies, a few studies have reported considering the instrument response time in deciding what averaging time to use for their logged data (L.Y. Chan et al., 2002; A.T. Chan et al., 2003), or attempted to develop an empirical equation or transfer function to improve the estimates of the true input concentrations (Larsen et al, 1965; Mage and Noghrey, 1972). However, the extent of error reduction using these methods has not been systematically examined.

Some researchers have used grab sampling in parallel with the real-time sampling as a quality assurance measure (Chan et al., 1999, 2002; Zagury et al., 2000; Esber et al., 2007). The grab sampling method uses a sampling bag to collect a single air sample over the whole real-time monitoring period, giving a time-integrated reference measurement to compare with the corresponding real-time average. However, for this method, a minimum sampling duration is required to collect an air sample sufficient for subsequent analysis, making it difficult to evaluate error for time scales of 60 s or less.

Zhang and Frey (2008) tested an empirical second-order exponential correction for reconstructing true input concentrations from simulated monitoring datasets. Their study also demonstrated a methodology to examine measurement errors for short time scales. However, their analyses were primarily based on datasets produced by computer simulation; errors for actual field data were not examined.

The purpose of this study is to develop a simple theoretical model that can reconstruct an accurate input concentration time series from logged measurements, based on the physical design of the electrochemical sensing device. Using experimental monitoring data, we test how well the model can estimate transient exposures to air pollutants occurring over seconds to minutes. In addition, errors corresponding to a wide range of averaging times are evaluated for actual measurements collected close to a point source,
to examine under what circumstances it will (or will not) be important to apply this reconstruction method for rapidly-varying measurements.

2.2 METHODOLOGY

Model derivation. When measuring close to a pollution source, the time scales over which the air pollution levels fluctuate are shorter than or comparable to the \( t_{50} \) monitor response time. Because the \( t_{50} \) response time of an electrochemical monitor is determined by the diffusion-limited design of the sensing cell (Warburton et al., 1998), the delay in monitor response for these conditions is expected to be primarily attributable to the diffusion properties of the sensor cell. To model the rate-limiting diffusional transport of an air pollutant from the sensor cell surface to the sensor cell interior, we use Fick’s First Law to describe the flux of the air pollutant entering or exiting the sensor cell:

\[
J = -D \nabla C = D \frac{(Y(t) - X(t))}{L_d}
\]  

For eq 1, \( J \) (\( \mu g \text{ cm}^{-2} \text{ s}^{-1} \)) is the diffusion flux; \( D \) (\( \text{cm}^2 \text{ s}^{-1} \)) is the diffusion coefficient of the air pollutant; and \( \nabla C \) (\( \mu g \text{ cm}^{-3} \text{ cm}^{-1} \)) is the concentration gradient. For this case, \( \nabla C \) can be expressed as the difference between the true input concentration, \( Y(t) \) (\( \mu g \text{ cm}^{-3} \)), and the concentration within the sensor cell – the monitor output measurement, \( X(t) \) (\( \mu g \text{ cm}^{-3} \)), divided by the length of the diffusion path, \( L_d \) (cm). Here, \( L_d \) can be treated as the equivalent thickness of the diffusion barriers in a monitor. By assuming a well-mixed condition for the sensor cell interior, we obtain a mass balance equation:

\[
\frac{dX(t)}{dt} V = \frac{AD}{L_d} (Y(t) - X(t))
\]  

For eq 2, \( V \) (\( \text{cm}^3 \)) is the interior volume of the sensor cell and \( A \) (\( \text{cm}^2 \)) is the cross-sectional area of the membranes. For a given monitor design, \( VL_d / AD \) is expected to be a constant that represents the time scale for diffusive transport in or out of the sensor cell.
By defining this expression as the time constant of the monitor, $\tau$ (in seconds), a model associating the true input concentration, $Y(t)$, with the monitor output measurement, $X(t)$, can be derived based on the monitor’s time constant:

$$\tau \frac{dX(t)}{dt} = Y(t) - X(t)$$ \hspace{1cm} (3)

This theoretical model (eq 3) is equivalent to an equation empirically determined by Larsen et al. (1965) from testing the dynamic response of 3 different types of continuous air sampling instruments. It is also consistent with the second-order exponential correction model used by Zhang and Frey (2008), if their higher order term is neglected.

**Model application.** Assuming a linear response within the measurable concentration range of a sensor, the derived model (eq. 3), a first-order ordinary differential equation, can be applied to (i) estimate the characteristic time constants of electrochemical monitors; (ii) reconstruct the true input concentration, $Y(t)$; and (iii) evaluate the errors of monitor output measurements, $X(t)$, corresponding to different averaging times. The mathematical and experimental methods used for these 3 applications, which are discussed in section 2.1 and 2.2, respectively, are summarized in Table 2.1, along with their primary results.
Table 2.1. Summary of the 3 types of monitor analyses performed in the present work

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<td></td>
<td>CO calibration gases applied to inlet of 38 CO monitors</td>
<td>Alternating high/low CO concentrations with varying frequency applied to inlet of 1 CO monitor</td>
<td>4 CO monitors placed at different distances from a indoor CO point source</td>
</tr>
<tr>
<td></td>
<td>Time constants were 36 s for forced rise and 44 s for unforced decay, on average.</td>
<td>Error in the monitor output measurement was reduced by 81% using the reconstruction method</td>
<td>Errors were below 15% for $T/\tau &gt; 10$, but in some cases above 50% for $T/\tau &lt; 1$</td>
</tr>
</tbody>
</table>

### 2.2.1 Mathematical Methods

**Characterization of monitor time constants.** To determine the time constant for an electrochemical sensor, one can set the input concentration in eq 3 to a constant value ($Y(t) = Y_c$), and assume that $X(t)$ and $Y(t)$ are initially in equilibrium ($X(0) = Y(0) = Y_o$), and then solve for the monitor output measurement, $X(t)$:

$$X(t) = Y_c \left[ 1 - \exp \left( -\frac{t}{\tau} \right) \right] + Y_o \exp \left( -\frac{t}{\tau} \right) \quad \text{for} \quad t \geq 0$$  \hspace{1cm} (4)

To estimate $\tau$ experimentally, one can make either $Y_c$ or $Y_o$ equal to 0 by using a zero gas to eliminate one of the two terms in eq 4. This will give a log-linear relationship between $X(t)$ and $t$, allowing $\tau$ to be estimated based on the slope of the regression line. For the case where the monitor is initially in equilibrium with the zero gas ($Y_o = 0$), and a span gas of concentration $Y_{span}$ is used to introduce a step increase in the input concentration ($Y_c = Y_{span}$), the rising monitor response can be described by the first term in eq 4.

Similarly, when the monitor is initially in equilibrium with the span gas ($Y_o = Y_{span}$), and a
zero gas is used to create a step decrease in the input concentration \((Y_c = 0)\), the decaying monitor response can be described by the second term in eq 4. Taking the natural logarithm of both sides of the two simplified equations, we obtain:

\[
\ln \left( \frac{Y_{\text{quan}} - X(t)}{Y_{\text{quan}}} \right) = \ln \left( 1 - \frac{t}{\tau_{\text{rise}}} \right) \quad \text{for} \quad t \geq 0
\]

(5a)

\[
\ln X(t) = \ln Y_{\text{quan}} - \frac{t}{\tau_{\text{decay}}} \quad \text{for} \quad t \geq 0
\]

(5b)

For eq 5a and 5b, \(\tau_{\text{rise}}\) and \(\tau_{\text{decay}}\) are the time constants that characterize the time response of the monitor for a step increase and a step decrease of input concentration, respectively.

**Reconstruction of actual input concentration.** To deduce the true input concentration, \(Y(t)\), for a monitor with a known time constant, \(\tau\), and the monitor output measurement, \(X(t)\), one can rearrange eq 3 to give:

\[
Y(t) = \tau \frac{dX(t)}{dt} + X(t)
\]

(6)

Hence, the true input concentration, \(Y(t)\), can theoretically be reconstructed based on the monitor’s real-time measurements and time constant. However, with a finite sampling frequency, determining \(dX(t)/dt\) may be challenging, because \(X(t)\) is not continuous in time. To estimate \(Y(t)\) from the discrete monitor measurements, we apply a finite difference method.

This numerical finite differencing approach involves using a finite step to approximate the first derivative of \(X(t)\) with respect to \(t\). Consider any 3 consecutive monitor output measurements, \(X_{n-1}\), \(X_n\), and \(X_{n+1}\) that are sampled at \(t_{n-1}\), \(t_n\), and \(t_{n+1}\), respectively. If \(X_n\) is not a sharp maximum or minimum, and the 3 data points form a reasonably smooth curve, then \(dX_n / dt\) can be approximated by eq 7, which is based on a commonly used
numerical analysis method, the Central Differencing Scheme (CDS) (Mathew and Fink, 2004):

\[
\frac{dX_n}{dt} \approx \frac{X_{n+1} - X_{n-1}}{t_{n+1} - t_{n-1}}, \quad n = 2, 3 \ldots (7)
\]

From eq 7 and eq 6, one can approximate \( Y_n \) based on the discrete real-time monitor output measurements and the time constant of the monitor:

\[
Y_n = \tau \left( \frac{X_{n+1} - X_{n-1}}{t_{n+1} - t_{n-1}} \right) + X_n, \quad n = 2, 3 \ldots (8)
\]

A certain degree of error may result when using eq 8 to estimate \( Y_n \), because a small level of electronic noise in the monitor can become influential when the response times of the monitor are relatively large or the logging intervals are very small. To reduce errors in the reconstructed input concentration time series (\( Y \)), one can further process measurements (\( X \)) with the Wiener filter (Dowling and Dimotakis, 1988) or the maximum entropy inversion (Lewis and Chatwin, 1995) methods to remove high-frequency components of \( X \) fluctuations due to monitor noise. In our study, we further applied curve smoothing to \( X_n \) reducing unwanted \( Y_n \) fluctuations attributable to this small amount of noise in \( X_n \).

\( \tau_{\text{rise}} \) and \( \tau_{\text{decay}} \) may not be exactly the same for a given monitor. The approximation of the first derivative of \( X_n \) with respect to \( t \) in eq 7 should be used with the appropriate value of \( \tau \), by computing \( Y_n \) using a piecewise algorithm:

\[
Y_n = \tau_{\text{rise}} \frac{X_{n+1} - X_{n-1}}{t_{n+1} - t_{n-1}} + X_n, \quad n = 2, 3 \ldots \text{ for } X_{n-1} < X_{n+1} \quad (9a)
\]

\[
Y_n = \tau_{\text{decay}} \frac{X_{n+1} - X_{n-1}}{t_{n+1} - t_{n-1}} + X_n, \quad n = 2, 3 \ldots \text{ for } X_{n-1} > X_{n+1} \quad (9b)
\]
\[ Y_n \approx X_n, \quad n = 1, 2, \ldots \quad \text{for} \quad X_{n-1} = X_{n+1} \quad (9c) \]

This piecewise algorithm can readily be carried out in many computer programming applications by using relational operators for comparing the relationship between \( X_{n-1} \) and \( X_{n+1} \) (Palm, 2005).

**Examination of monitoring errors for different averaging times.** The calculation of a time-averaged monitor output measurement, \( \overline{X(t)} \), may incorporate one or more rise and decay cycles. Since both the rise and decay are subject to the slowing effect of response time, the instrument’s underestimate of \( Y(t) \) in the rise periods can be compensated by its overestimate of \( Y(t) \) in the decay periods. Thus, the larger the averaging time, the more the error of each time-averaged monitor output measurement, \( \overline{X(t)} \), can be minimized. This time-averaged error can be evaluated mathematically by integrating eq. 3 over an averaging time, \( T \):

\[
\int_{t_i}^{t_i+T} Y(t) \, dt - \int_{t_i}^{t_i+T} X(t) \, dt = \tau \int_{t_i}^{t_i+T} \frac{dX(t)}{dt} \, dt, \quad i = 1, 2, \ldots \quad (10)
\]

For eq 10, \( t_i \) represents the initial time of the \( i \)th time interval. When integrated and divided by \( T \) to obtain the time average expression of each term in eq 10, this gives:

\[
\overline{Y_i} - \overline{X_i} = \frac{\tau}{T} \left[ X(t_i + T) - X(t_i) \right], \quad i = 1, 2, \ldots \quad (11)
\]

In eq 11, \( \overline{Y_i} \) is the \( i \)th time-averaged true input concentration and \( \overline{X_i} \) is the \( i \)th time-averaged monitor output measurement. As shown above, the difference between \( \overline{X_i} \) and \( \overline{Y_i} \) is directly proportional to the difference between the final and initial monitor output concentrations multiplied by the ratio of \( \tau \) to \( T \). In other words, the error in \( \overline{X_i} \) is expected to depend on how rapidly the actual concentration varies with time and what corresponding \( \tau / T \) is chosen. This indicates that for typical long-term exposure
assessment (i.e. weekly averages) where $T \gg \tau$, errors in time-averaged monitor output measurements are expected to be very small.

The dimensionless ratio ($\tau / T$) found in eq 11 has been previously proposed as an important indicator for evaluating the time-response error of electrochemical sensors, based on a mathematical derivation for a single step increase of concentration (Esber et al., 2007). An additional insight from eq 11 is that this dependence of the monitoring error on $\tau / T$ is expected to also apply to more complicated concentration fluctuations, with multiple cycles of exponential rise and decay in the monitor output measurements. Ott et al. (1994) implemented this relationship to model the smoothing effect of the motor vehicle’s passenger compartment on the time-varying CO concentrations entering from moving traffic.

2.2.2 Experimental Methods

**Characterization of monitor time constants.** To provide data for examining the time response characteristics of electrochemical monitors, we performed controlled experiments using 38 Langan CO sensors (Langan Products, Inc., San Francisco, CA, USA). In these experiments, we initially applied a CO span gas ($Y_{\text{span}} = 50$ or $60$ ppm, Scott Specialty Gases, Inc., Plumsteadville, PA, USA) to create a rising monitor response. After an equilibrium was reached (at the maximum value, $X_{\text{max}}$), we applied either ambient air or zero calibration gas (0.3 ppm, Scott Specialty Gases, Inc., Plumsteadville, PA, USA) to produce a decay response. Figure 2.1 shows an example of monitor output measurements for a step increase at $t = 0$, using span gas, followed by a step decrease at $t = t_a$, using zero gas. In this controlled experiment, monitors were set up to measure CO concentrations at a 2-s time resolution. For each monitor, $\tau_{\text{rise}}$ was determined by a log-linear regression between $Y_{\text{span}} - X(t)$ and $t$ for $0 < t \leq t_a$, and $\tau_{\text{decay}}$ was determined by a log-linear regression between $X(t)$ and $t$ for $t > t_a$.

Two different types of monitor response times were evaluated: (i) “forced $\tau$”, representing the forced flow of a calibration gas into the monitor, and (ii) “unforced $\tau$”, representing the natural diffusion of ambient air into the monitor. A pressurized cylinder
of calibration gas was attached by tubing to a small cap that is placed over the sensor intake, causing the concentration to rise in a forced manner as the gas flowed to the sensor cell. Exposing the sensor to open air with no span gas (unforced) was expected to be different from attaching a pressurized cylinder with a zero gas (forced).

**Figure 2.1.** An example of monitor output measurements, $X(t)$, for a step increase ($t = 0$) and a subsequent step decrease ($t = t_a$) of CO input concentrations, $Y(t)$, used to determine monitor rise and decay time constants $\tau_{\text{rise}}$ and $\tau_{\text{decay}}$, respectively. $Y_{\text{span}}$ is the CO concentration of span gas. $Y_{\text{zero}}$ is the CO concentration of zero gas or ambient air. $t_a$ is the time when the zero gas was introduced in place of the span gas. $X_{\text{max}}$ is the monitor output measurement at $t = t_a$, the maximum monitor reading reached. $t_{50}$ and $t_{90}$ are the times required for a sensor to read 50% and 90%, respectively, of its final value.
**Reconstruction of actual input concentration.** To test our finite difference method (eq 9a-9c) for reconstructing the true input concentrations, we conducted an experiment using CO calibration gases as controlled sources. In this test, a monitor with forced $\tau_{\text{decay}} = 36.6$ s and forced $\tau_{\text{rise}} = 31.5$ s was set up to measure concentrations every 2 s over a 23-min period. Tubing was used to connect the monitor with 50 ppm and 0.3 ppm CO calibration gases alternately several times at each of 3 different frequencies (with input durations of 146 s, 42 s, and 12 s, respectively). This approach created a series of known repeated step inputs to compare with the reconstructed concentrations (Figure 2.2a).

The Smoothing Spline fitting method (Dierckx, 1995) in MATLAB Curve Fitting Toolbox was first used to generate a new set of monitor output measurements with the same 2-s time resolution, reducing the electronic noise of the monitor for the subsequent finite differencing computation. The $R^2$ of this fitting process was set to 0.999, which corresponds to 0.007 for the smoothing parameter.

**Examination of monitoring errors for different averaging times.** To examine the magnitudes of the errors in the time-averaged monitor output measurements, $\bar{X}(t)$, for different averaging times, $T$, and different variations in the input concentrations, we carried out an indoor CO experiment in a California single family house. We placed a CO point source at the center of a $9 \times 4 \times 3$ m living room to release 99% CO (Scott Specialty Gases, Inc., Plumsteadville, PA, USA) at a constant emission rate controlled by a Model 5896 mass-flow controller (Emerson Electric, Co., St. Louis, MO, USA). The air flow rate was calibrated using a Gilibrator Primary Flow Calibrator (Sensidyne, Inc., Clearwater, FL, USA). In this indoor experiment, the controlled air flow rate (12.9 cc/min) gave an equivalent source emission rate of 14.8 mg/min.

To measure differing extents of concentration variation, 3 monitors were set up along one direction at horizontal distances of 0.25 m, 2 m and 4 m from the source, and an additional monitor was placed in the opposite direction at 0.25 m from the source. Both the source and the monitors were mounted on tripod stands at the same elevation (1 m from the ground). The monitors with unforced $\tau_{\text{decay}} = 32-47$ s simultaneously recorded
concentrations every 15 s over a 1-h sampling period. Since $\tau_{\text{rise}}$ values were determined by forcing the span gas into each of the sensors, the unforced $\tau_{\text{rise}}$ values remained unknown. To reconstruct $Y(t)$ for this real-world unforced CO experiment, the unforced $\tau_{\text{rise}}$ was assumed to be the same as the unforced $\tau_{\text{decay}}$ for each of the 4 monitors.

For averaging times, 9 values of $T$ ranging from 15-3600 s were considered. To compare the average errors among these time resolutions, the mean absolute relative error was used:

$$\text{Error} = \frac{1}{n} \sum_{i=1}^{n} \left| \frac{\bar{X}_i - \bar{Y}_i}{\bar{Y}_i} \right|,$$

$$i = 1, 2, \cdots$$

In eq 12, $n$ is the number of time-averaged concentrations (total sampling period/averaging time interval), and $\bar{X}_i$ and $\bar{Y}_i$ are the $i$th time-averaged monitor output measurement and reconstructed input concentration, respectively.

**Monitor calibration.** Before the start of each experiment, CO monitors were initially exposed to ambient air to adjust the monitor reading to background level (~0.5 ppm) for the zero calibration. After that, they were connected via tubing to the 50 ppm or 60 ppm span gas cylinders for 3-5 minutes. Once equilibrium was reached, each monitor reading was adjusted to match the appropriate span gas concentration. Sensors showing any unstable digital reading during the two-point calibration procedure were excluded from our experiments.

**2.3 RESULTS AND DISCUSSION**

**2.3.1 Characterization of Monitor Time Constants**

We evaluated both the rise and the decay periods for 38 Langan CO monitors that had been purchased over a number of years. Six of the older monitors either had time constants that differed from the cohort average by > 2 standard deviations, or had $R^2 < 0.80$ for one or both of the log-linear regressions. The newer 32 monitors had average
forced $\tau_{\text{rise}}$ values of 36 s (± 5 s), and unforced $\tau_{\text{decay}}$ values of 44 s (± 8 s). More than 96% of the log-linear regression lines (62/64) had $R^2$ values above 0.90.

For 84% of the monitors (27/32), larger $\tau$ values were measured for the unforced decay period than for the forced rise period; on average, $\tau_{\text{decay}} - \tau_{\text{rise}}$ was 10 s (± 5 s). Since the CO sensor works via the principle of diffusion, we hypothesized that the forced flow of CO span gas in our experiments was reducing the $\tau_{\text{rise}}$ values. Via subsequent testing of 3 monitors, we found that the $\tau_{\text{decay}}$ values decreased when the zero gas was forced, and that the $\tau_{\text{rise}}$ and $\tau_{\text{decay}}$ values more closely matched each other when both were forced.

The most appropriate $\tau$ value to use in modeling the behavior of the CO sensor in the field is the $\tau$ found during the unforced decay period (36-52 s), since exposures to environmental CO levels are unforced.

### 2.3.2 Reconstruction of Actual Input Concentration

Figure 2.2(a) shows the resulting monitor measurements when applying a series of repetitive step inputs to a sensor. As the frequency of the step inputs increased, the discrepancy between the monitor output measurement, $X(t)$, and the true input concentration, $Y(t)$, became more pronounced, due to the non-instantaneous response of the monitor. This result illustrates that significant errors would be expected when measuring rapidly varying input concentrations.

Using the real-time monitor output measurements and our estimates of the forced time constants for the monitor, we modeled the reconstructed input concentration, $Y(t)$, using the finite difference method. Figure 2.2(b) shows that this simple numerical method can reconstruct different durations of step inputs (146 s, 42 s, and 12 s) with reasonable accuracy. The mean absolute difference between the reconstructed and the true input, $Y(t)$, was 3.25 ppm ($n = 676$), while the corresponding mean absolute difference between the monitor output, $X(t)$, and the true input, $Y(t)$, was 16.94 ppm. Thus, the reconstruction method resulted in an overall error reduction of 81%, on average, for 2-s readings. These results indicate that the first order model used here can closely
reconstruct the true input concentration for the range of input frequencies considered in the experiment, without needing the higher order correction term used by Zhang and Frey (2008).

We do see fluctuations in $Y(t)$ at the constant-concentration levels of both 50 ppm and 0.3 ppm. Monitor electronic noise embedded in $X(t)$, though imperceptibly small in Figure 2.2(b), is likely to cause fluctuations in the reconstructed $Y(t)$ for time periods when input concentrations remain constant.

**Figure 2.2.** (a) Time series for 3 different frequencies of repetitive span-gas step inputs, $Y(t)$, applied to a CO sensor (with input durations of 146 s, 42 s, and 12 s, respectively), and their corresponding monitor output measurements, $X(t)$. Both $X(t)$ and $Y(t)$ are at 2-s time resolution. (b) $Y(t)$ reconstructed by the finite difference model based on the monitor output measurements, $X(t)$, and the time constants of the monitor (forced $\tau_{\text{decay}} = 36.6$ s and forced $\tau_{\text{rise}} = 31.5$ s). Both $X(t)$ and $Y(t)$ are at 2-s time resolution.
2.3.3 Examination of Monitoring Errors for Different Averaging Times

Using data from the experiment where 4 monitors were set up at different distances from a uniformly emitting CO point source, Figure 2.3(a) shows a 25 min time series of one monitor’s 15-s output measurements, $X(t)$, (with $T / \tau = 0.32$) alongside the corresponding reconstructed 15-s true input concentrations, $Y(t)$. Here, the reconstructed $Y(t)$ appears smoothly varying even without applying curve smoothing for $X(t)$. As this example shows, when rapidly varying concentrations occurred (0.25 m from the source), the discrepancy between the monitor output measurement, $X(t)$, and the reconstructed true input concentration, $Y(t)$, was often significant due to the non-instantaneous response of the monitor ($\tau = 47$ s). This result indicates that to capture the magnitude of transient exposures to rapidly varying concentrations more accurately, it is advisable to use the reconstruction model. Based on the same data set, Figure 2.3(b) and 2.3(c) show the comparisons between the time-averaged monitor output measurements, $\overline{X(t)}$, and the corresponding time-averaged values of the reconstructed input concentrations, $\overline{Y(t)}$, when 75-s and 150-s averaging times (with $T / \tau = 1.6$ and 3.2) are used. Here, $\overline{X(t)}$ and $\overline{Y(t)}$ more closely converge as the averaging time increases, reducing the discrepancy between the monitor output measurements and the true input concentrations. By choosing a sufficiently large averaging time, $\overline{X(t)}$ can become a reasonable approximation for $\overline{Y(t)}$, making input signal reconstruction unnecessary.
Figure 2.3. Comparison between monitor output measurements (at 0.25 m from an indoor CO point source) and input concentrations reconstructed by the finite difference model, at 3 different averaging times: (a) 15-s monitor output measurements, \( X(t) \) (\( T/\tau = 0.32 \)), versus 15-s reconstructed input concentrations, \( Y(t) \). (b) 75-s time-averaged monitor output measurements, \( \bar{X}(t) \) (\( T/\tau = 1.6 \)), versus 75-s time-averaged values of reconstructed true input concentrations, \( \bar{Y}(t) \). (c) 150-s time-averaged monitor output measurements, \( \bar{X}(t) \) (with \( T/\tau = 3.2 \)), versus 150-s time-averaged values of reconstructed input concentrations, \( \bar{Y}(t) \).
Figure 2.4 shows the mean absolute relative error (see eq 12) versus the corresponding dimensionless ratio, $T/\tau$, for each of the 4 monitors over a wide range of time scales, each utilizing the same 1-hr measurement period. As a measure of the relative variability for these 4 sets of environmental measurements, the coefficient of variation (CV) — the ratio of the standard deviation to the mean — was calculated for each set of monitor output measurements (CV with respect to $X(t)$). For a larger CV, there was greater error in the monitor measurements, especially for $T/\tau < 2$. This result is consistent with Zhang and Frey (2008), who plotted 4 different frequencies/time scales of input fluctuations, and found that as the frequency of the input signal increases, the errors in monitor output measurements became more pronounced.

As $T/\tau$ increased, the mean absolute relative error decreased for these 4 data sets, dropping below ~15% for $T/\tau > 10$. The 2 monitors located closest to the source (0.25 m) had the largest CV’s (1.34 and 0.85) due to more dramatic fluctuations in the actual input concentrations. They showed mean absolute relative errors above 50% for $T/\tau < 1$.

Esber et al. (2007), who used grab sampling to evaluate continuous CO measurements of vehicle-related emissions, showed that errors in trip-averaged exposures were within a range of 0.6-15% for ratios of overall averaging times to the monitor time constant ranging from 45 down to 15, respectively. Our experimental results, which cover a much larger range of $T/\tau$ values, are comparable within this range, with $T/\tau$ values of 56 to 14 corresponding to errors of 0.6-11%. These findings indicate that by selecting an averaging time that is sufficiently large compared with the monitor time constants, accurate estimates of $\bar{Y}(t)$ can be obtained directly, without needing to use the reconstruction algorithm. But as $T/\tau$ decreases, the error increases noticeably, especially for rapidly varying concentrations with large CV values. Thus, for situations where transient spikes in concentration are important to quantify accurately and the desired averaging time is less than or comparable to the monitor’s time constant, the reconstruction method should be used.
Figure 2.4. Mean absolute relative errors (“Error”) between the time-averaged monitor output measurements, $X(t)$, and the time-averaged values of the reconstructed input concentrations, $Y(t)$, with respect to different ratios of averaging time to monitor time constant, $T/\tau$ for 4 different amounts of input concentration variations. Concentration variations were quantified using the coefficient of variation (CV) — the ratio of standard deviation to mean for each set of 15-s monitor output measurements, $X(t)$, over a 1-h sampling period ($n = 241$).
2.4 SUMMARY AND IMPLICATIONS

This study shows that Fick’s First Law of diffusion provides a theoretical basis for the behavior of typical electrochemical sensors. Using this principle, we implemented a first-order model to: (i) determine the characteristic time constants of the monitors; (ii) reconstruct the true input concentration time series; and (iii) estimate monitor errors for different averaging times. Using this model we found:

- The experimentally determined time constants of 38 tested electrochemical CO monitors ranged from 30 to 50 s. These characteristic time constants were used, with the model, to reconstruct a rapidly varying input concentration time series with reasonable accuracy.

- By choosing an averaging time that is sufficiently large compared with the monitor time constants, the measurement error for a rapidly-varying concentration time series can be minimized effectively. We found that, when the ratio of the overall averaging time to the monitor's time constant was greater than 10, the mean absolute relative error dropped below ~15% over a wide range of variations in the input concentration.

- Reconstructing the true input concentrations from a monitor’s output measurements should be performed when the exposure time scales of interest are less than or comparable to the time constant of the monitor (e.g. acute exposure to toxic gases).

When pollutant fluctuations are pronounced and rapid, and transient exposures to air pollutants are of interest, it becomes important to consider the delay in sensor’s time response when interpreting real-time monitoring datasets. The reconstruction method we present in this study is expected to be applicable to real-time gas sensors involving diffusion-limited electrochemical reactions, regardless of which air pollutant is being measured. It may be helpful for investigators wishing accurately to measure rapidly-varying concentrations on the order of seconds to minutes, such as measurements extremely close to an active point source or near moving traffic.
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REFERENCES


CHAPTER 3

Modeling Exposure Close to Air Pollution Sources in Naturally-Ventilated Residences: Association of the Turbulent Diffusion Coefficient with Air Change Rate


ABSTRACT

In indoor models of exposure close to a source, an isotropic turbulent diffusion coefficient is used to represent the spread of time-averaged concentration in the presence of an indoor air pollution point source. However, the magnitude of this parameter has been difficult to assess experimentally for indoor spaces due to limitations in the number of monitors available to capture the concentration field over the entire indoor space. Up to 37 real-time monitors were used to measure simultaneously CO at different angles and distances from a continuous indoor point source, during initial 30-min tracer gas releases. We conducted 11 experiments to assess turbulent diffusion coefficients in 2 residential houses, systematically varying window openings to create natural ventilation conditions ranging from <0.2 to >5 air changes per h. An eddy diffusion model was used, along with 30-min time-averaged concentrations at each position, to determine the turbulent diffusion coefficients, which ranged from 0.001 to 0.015 m$^2$ s$^{-1}$, reproducing the observed concentration proximity profiles with reasonable accuracy over the full extent of radial distances. In addition, a significant linear correlation was seen between simultaneously measured air change rates and turbulent diffusion coefficients in each indoor space. To combine data from the 2 indoor spaces, which had different dimensions, each turbulent diffusion coefficient was normalized by dividing by the square of a length scale representing the room volume, to create an air mixing rate (h$^{-1}$). The resulting

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2 A version of this chapter has been submitted to *Environmental Science and Technology*
relationship between air mixing rates and air change rates for the 2 indoor spaces showed a significant overall positive linear correlation \((R^2 = 0.94)\). This result suggests that the indoor turbulent diffusion coefficient could be estimated using two readily-measurable parameters: the air change rate and the room dimensions. The present characterization of the turbulent diffusion coefficient is useful for modeling the spatial variability of exposures in close proximity to an active indoor air pollution source, such as a cigarette smoker and other household source emissions.

3.1 INTRODUCTION

Typically, human exposure to an indoor emission source has been estimated by the well-mixed mass balance model (e.g., Shair and Heitner, 1974; Hayes, 1991; Keil, 1998; Burke et al., 2001; Ott et al, 2003; von Grote et al, 2003; Vernez et al, 2006). This model assumes that soon after being emitted from a point source, air pollutants become instantaneously, completely well-mixed. Therefore, the concentrations are represented as spatially homogeneous within an indoor space, but varying with time due to emission and removal pathways for the pollutants. The indoor time scale for approaching a well-mixed state following release is typically < 1 h (Baughman et al, 1994; Drescher et al, 1995; Klepeis, 1999), so this modeling approach can provide a simple and accurate assessment of the long-term exposure to short-duration indoor emissions, provided that the source emission and mixing time scales are much smaller than the duration over which the time-averaged concentration (the estimate of exposure) is considered.

However, for a source that releases air pollutants over a duration comparable to the exposure time of interest, imperfect mixing during the emission period becomes important to consider. During this active source period, exposures in close proximity to the active source are expected to be substantially higher than those further away from it, and this “proximity effect” will not be captured by a uniform mixing model (Rodes et al, 1991; Furtaw et al., 1996; McBride et al., 1999; McBride 2002; Ott et al, 2002; Ferro et al., 1999, 2004, 2009; Acevedo-Bolton et al., 2010).
To model the effect of proximity on personal exposure, various mass transfer models have been proposed and used that invoke the concept of isotropic turbulent diffusion to describe the mixing of an indoor air pollution point source. Analytical solutions of Fick’s Law of diffusion have been used to describe turbulent mixing of indoor air and to model exposures at different distances from a continuous point source (Wadden et al., 1989, Conroy et al., 1995, Demou et al., 2009) and from a short-duration source (Drivas et al., 1996). For the case of forced air flow in an indoor environment, Scheff et al. (1992) used an advection-diffusion equation to describe the transport of air pollutants from 2 indoor point sources and predicted exposures at different indoor positions of a room. More detailed summaries and discussions of these models have been presented by Keil (2000) and Jayjock et al. (2007). In addition to these analytical approaches, Nicas (2001) invoked the theory of random walk to describe turbulent diffusion transport indoors and implemented it in a stochastic Markov Chain model to consider the spatial variations of exposure in indoor environments.

All these models rely on one empirical parameter—the isotropic turbulent diffusion coefficient (\(K\))—to describe how fast the spatial spread of air pollutants (symmetrical with respect to the source) increases with time due to turbulent mixing indoors. Field determination of \(K\) for an indoor space of interest typically involves simultaneous concentration measurements at different indoor positions.

One approach for finding \(K\) begins by defining the mixing time as the time at which the coefficient of variation (CV) of concentrations measured at all monitoring points drop permanently below 10%. Baughman et al. (1994) and Drescher et al. (1995) empirically determined mixing times of a pulse release of a tracer gas in a controlled experimental room under natural and forced ventilation conditions. Klepeis (1999) analyzed spatial concentration data of cigarette smoke from a bedroom and a tavern to characterize mixing times, using similar method. This characterization of indoor mixing times allows the use of eddy diffusion models to optimize/determine a \(K\) that gives comparable mixing times.
As another approach, $K$ can be estimated by fitting the model predictions to measurements at different distances from the source. Scheff et al. (1992) measured 1-h time-averaged concentrations at 9 indoor positions in a mechanically-ventilated manufacturing facility, and used the least square method to determine $K$ as well as 3 other unknown variables in an advection-diffusion model. Conroy et al (1995) placed 2 monitors along an axis at 2 different distances from a chemical emission source using 1-h time-averaged concentrations and the solution of Fick’s second law to solve for $K$ as well as the source emission rate in an electroplating shop. Demou et al (2009) used 2 instantaneous measurements of a monitor, along with the mass emission rate of an indoor point source, to determine $K$ in a vehicle repair shop.

As summarized above, past efforts to characterize indoor $K$ values have considered only large occupational environments and have been limited by how few monitors were available to capture the spread of source emissions over the entire indoor space. In addition, given that these studies were conducted in spaces of different-size, under a broad range of ventilation and thermal conditions with reported $K$ values spanning 2 orders of magnitude (Keil, 2000), it is difficult to know which value of $K$ is the most appropriate to select for another indoor space of interest.

To generalize $K$ under different indoor conditions, Karlsson et al. (1994) derived a turbulence dissipation equation based on the conservation of turbulent energy, leading to a model that associates $K$ with the air change rate ($ACH$) of a room, the air velocity at ventilation intake, and the temperature gradient between ceiling and floor (Drivas et al. 1996). This theoretical model can yield $K$ based on the specific ventilation and thermal conditions in a room. However, the estimated value has been found to be several orders of magnitude higher than $K$ values measured experimentally, and it is applicable only for mechanically ventilated indoor spaces (Demou et al., 2009).

The first goal of our study was to determine accurate estimates of $K$ in a type of indoor environment that has not previously been evaluated: naturally-ventilated residential homes. We used a real-time CO monitoring array (up to 37 monitors) to simultaneously
monitor throughout the indoor space during a series of controlled CO tracer gas releases. Using our estimated $K$ values, we quantitatively tested how well the isotropic eddy diffusion formulation used in the indoor proximity exposure models can describe exposures at different distances from an air pollution point source in real-world indoor residential settings. Our second goal was to examine how the air change rate and room size affect the magnitude of the turbulent diffusion coefficient. We conducted a series of 11 experiments with factorial designs in 2 houses under a range of natural ventilation rates to explore whether $K$ can be estimated using readily-measured parameters like the air change rate and room dimensions.

3.2 METHODOLOGY

3.2.1 Experimental Method
This study involved 2 rooms, each in a different house in northern California (Figure 3.1). The first indoor space (Room #1) is a 9.4 × 4.1 m living room with a single-sloped ceiling, in a two-story occupied single family home in Redwood City. The second (Room #2) is a 5.6 × 4.4 m family room with a double-sloped ceiling, in a one-story occupied ranch style home in Watsonville. In each indoor space, a CO tracer gas point source placed at the center of the room released 99.99% CO (Scott Specialty Gases, Inc., Plumsteadville, PA, USA) at a flow rate of ~20 cc min$^{-1}$ (~400 µg s$^{-1}$ at 25°C, 1atm), controlled by a Model 5896 mass-flow controller (Emerson Electric Co., St. Louis, MO, USA). The air flow rate was calibrated and monitored using a Gilibrator Primary Flow Calibrator (Sensidyne, Inc., Clearwater, FL, USA). Further details on the full indoor tracer study are available in Acevedo-Bolton et al. (2010).

The initial 30-min tracer gas release duration was chosen to provide a sufficient averaging time to account for the random fluctuations of CO concentration due to turbulent mixing indoors. This time period is analogous to the 10-30 min averaging times used in the Pasquill-Gifford curves for plume dispersion outdoors (Gifford, 1961).

To examine the spatial spread of CO for this indoor tracer gas release, we deployed 37 (Room #1) and 30 (Room #2) real-time CO monitors (Langan Products, Inc., San
surrounding the source at different radial distances and angles, measuring CO concentrations every 15 s. Figure 3.1(a) and 3.1(b) show a plan view of the array configuration in Room #1 and Room #2 of each house, respectively, along with the source position at the intersection of two defined perpendicular axes (x- and y- axes). For each room, 16 monitors were deployed in close proximity along the two axes: 4 each at 0.25, 0.5, 1, and 2 m from the source. To better capture the transient directional transport of CO in close proximity, additional monitors were placed at 1 and 2 m from the source, giving a monitor angle spacing of 30 degrees for Room #1, and 45 degrees for Room #2, respectively. Along the x-axis of Room #1, we placed 4 monitors at distances of 3 and 4 m, and 1 monitor at 5 m from the source. In Room #2, we added 2 monitors at 2.8 m from the source along the x-axis, and 4 monitors at 4 corners of the rectangular room (3.56 m from the source). The source and monitors were all placed at 1.0 m height, chosen to approximate an adult’s breathing height while sitting.
Figure 3.1. Plan view of CO monitoring array configurations in the 2 rooms studied (Room #1 and Room #2) in 2 residential houses. The filled circles show the positions of CO monitors surrounding the CO point source (unfilled star) located at the intersection of the two perpendicular axes (x- and y-axes).

As shown in Table 3.1, the factorial experimental design involved varying the window positions and the number of windows open to create a range of natural ventilation settings, categorized as state 0 through state 3 or 4. The air change rate ($ACH$) for each experiment was measured using Sulfur Hexafluoride ($SF_6$) gas (Scott Specialty Gases, Inc., Plumsteadville, PA, USA). $SF_6$ was released for 10-20 min at the center of the room at the beginning of each experiment, with 2 real-time $SF_6$ monitors (Brüel Kjaer, Inc., Nærum, Denmark), placed at the 2 ends of the x-axis in each room to measure $SF_6$ concentration every 1 min over ~4 h. After the end of the release period, once $SF_6$ became well-mixed in the room, an indoor mass balance equation (eq. 1) was applied to model the decreasing $SF_6$ gas concentration with time (Howard-Reed et al., 2002).
For eq 1, $C_{SF_6}(t)$ (ppm) is the concentration of SF$_6$ at time $t$, $t_o$ (min) is the time at which SF$_6$ becomes well mixed in the indoor environment, and $ACH$ (min$^{-1}$) is the air change rate. Using this equation, the $ACH$ of each experiment can be found from the slope of the log linear regression between $C_{SF_6}(t)$ and $t$. The $R^2$ value of the regression line reflects how reliable the estimated $ACH$ is.

Table 3.1 Opening width of windows in the 2 houses for different natural ventilation settings (state 0 - state 3 or 4) used in the factorially designed experiments.

<table>
<thead>
<tr>
<th>Window ID #</th>
<th>Window Location</th>
<th>Nature Ventilation Setting</th>
<th>Opening width (Percent of Opening) $^{(2)}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>State 0  State 1  State 2  State 3  State 4</td>
</tr>
<tr>
<td>House #1</td>
<td></td>
<td></td>
<td>State 0  State 1  State 2  State 3  State 4</td>
</tr>
<tr>
<td>#1</td>
<td>Den (1$^{st}$ floor)</td>
<td>0''(0%)  4''(18%)  8'' (36%)  22''(100%)</td>
<td></td>
</tr>
<tr>
<td>#2</td>
<td>Master bedroom (2$^{nd}$ floor)</td>
<td>0''(0%)  4''(20%)  8'' (40%)  20''(100%)</td>
<td></td>
</tr>
<tr>
<td>#3</td>
<td>2$^{nd}$ bedroom (2$^{nd}$ floor)</td>
<td>0''(0%)  6''(30%)  12''(60%)  20''(100%)</td>
<td></td>
</tr>
<tr>
<td># of experiment</td>
<td></td>
<td></td>
<td>1          2          1          1</td>
</tr>
<tr>
<td>House #2</td>
<td></td>
<td></td>
<td>State 0  State 1  State 2  State 3  State 4</td>
</tr>
<tr>
<td>#1</td>
<td>Family room$^{(1)}$</td>
<td>0''(0%)  0'' (0%)  0''(0%)  8''(50%)  16''(100%)</td>
<td></td>
</tr>
<tr>
<td>#2</td>
<td>Family room$^{(1)}$</td>
<td>0''(0%)  3''(19%)  8''(50%)  8''(50%)  16''(100%)</td>
<td></td>
</tr>
<tr>
<td>#3</td>
<td>Kitchen</td>
<td>0''(0%)  0''(0%)  8''(29%)  8''(29%)  16''(57%)</td>
<td></td>
</tr>
<tr>
<td># of experiment</td>
<td></td>
<td></td>
<td>1          1          1          2          1</td>
</tr>
</tbody>
</table>

$^{(1)}$ Family room is the sampling room (Room #2) where we deployed CO and SF$_6$ monitors.
$^{(2)}$ Percent of opening was calculated by $100\% \times ( \text{opening width/fully-open width} )$ for each window.

We used 2 digital Hygro-Thermometers (Sunleaves Inc., Bloomington, IN, USA) to simultaneously record temperatures near the ceiling and the floor in each room (3 m apart in Room #1 and 2.3 m apart in Room #2), before and after some of the experiments, providing vertical temperature gradients as an indication of the magnitude of indoor thermal stratification. We placed a 2-D ultrasonic anemometer (WindSonic™ Model, Gill,
Inc., Hampshire, England) outdoors close to each house (1.5 m from the ground) to measure wind speed and direction every s.

3.2.2 Quality Assurance for CO Monitor Array Measurements.

Monitor calibration. Before the start of each experiment, CO monitors were initially exposed to ambient air, adjusting the monitor reading to background level (0.5 ppm) for the zero calibration. Then they were connected via tubing to the 50 ppm or 60 ppm NIST traceable span gas cylinder for 3–5 min. Once equilibrium was reached, the span potentiometer on each monitor was adjusted to match the appropriate calibration gas concentration. Sensors showing any unstable digital reading during the 2-point calibration procedure were excluded from the experiment.

Averaging time. Based on a previous study (Cheng et al, 2010), the response times of CO monitors used in our experiments ranged from 30-50 s, giving monitoring errors < 15% for an averaging time > 10 min. For the 15-s monitor array measurements in this study, we computed the time-averaged concentrations over the 30-min duration of each experiment (averaging time of 30 min) to minimize the monitoring bias due to the non-instantaneous time response of the monitors to fluctuations in concentration.

3.2.3 Characterization of Turbulent Diffusion Coefficient

To estimate the turbulent diffusion coefficient ($K$) that can be applied to the currently available indoor eddy diffusion models, we followed the general assumption/simplification in those models of negligible mean/time-averaged indoor advection under natural ventilation conditions, and invoke Fick’s Second Law of diffusion to describe the CO concentration field over the experimental duration (30 min). We also assume, for a source at the center of the room during this initial 30 min of emission, that the removal of CO from the room due to air exchange can be neglected without loss of accuracy in determining $K$. This assumption was made because, for our natural and low ventilation settings, the time scales of pollutant removal for most of our experiments (>2 h) are larger than the 30-min experimental duration, making the loss of CO negligible over the time scale (30 min) of interest. For a continuous source, CO
concentration as a function of time, $t$ (s) and radial distance from the source, $r$ (m) can be described as in Crank (1975):

$$C(r, t, K) = \frac{q}{4\pi K r} \left( 1 - \text{erf} \left( \frac{r}{\sqrt{4Kt}} \right) \right) \quad (2)$$

For eq 2, $C$ ($\mu g/m^3$) is the CO concentration; $q$ ($\mu g/s$) is the mass emission rate of CO; $K$ ($m^2/s$) is an isotropic indoor turbulent diffusion coefficient; and erf is the Error Function ranging from 0 to 1 (Charbeneau, 2000) and is related to the integral of the normal distribution. Wadden et al. (1989), Conroy et al. (1995), and Demou et al. (2009) have applied eq 2 to experimental datasets to characterize $K$ in different occupational workplaces. Bennett et al. (2003) used the steady state form of eq 2 to determine $K$ for a small embalming room, based on a dataset generated via CFD simulation.

To account for the reflection of CO from wall surfaces, one can express eq 2 in the Cartesian coordinate system, and introduce “image sources” with respect to each wall plane – hypothetical sources used to satisfy no-flux boundary condition at walls. Drivas et al. (1996) has employed this method to model the air pollutant reflection from 6 walls of a rectangular room, using infinite series of image sources. Given the short-term experimental duration (30 min), image sources relatively far away from the indoor region of interest can be neglected. Thus, in addition to the real CO source, we introduce 6 image sources closest to the indoor space (one image source for each wall boundary) to create eq 3 as our model equation.

$$C_{\text{model}}(x, y, z, t, K) = \frac{q}{4\pi K} \left\{ \frac{1-\text{erf}(\sqrt{\frac{(x-x_o)^2+(y-y_o)^2+(z-z_o)^2}{4Kt}})}{\sqrt{(x-x_o)^2+(y-y_o)^2+(z-z_o)^2}}} \right\} + \sum_{i=1}^{6} \frac{1-\text{erf}(\sqrt{\frac{(x-x_i)^2+(y-y_i)^2+(z-z_i)^2}{4Kt}})}{\sqrt{(x-x_i)^2+(y-y_i)^2+(z-z_i)^2}}} \quad (3)$$

For eq 3, $C_{\text{model}}$ is CO concentration modeled by superposing the real CO source at $(x_o, y_o, z_o)$ with 6 image sources at $(x_i, y_i, z_i)$. By defining the positions of 6 wall boundaries as $x$
= x_{wall1}, x = x_{wall2}, y = y_{wall1}, y = y_{wall2}, z = z_{wall1}, and z = z_{wall2}, the coordinates of the 6 image sources can be determined (see Table 3.2). Due to the more complicated geometry of the ceiling in the two studied rooms (a single-sloped ceiling and a double-sloped ceiling), we simplified the approach by assuming an average ceiling height for reflection, calculated as the mean of the maximum and minimum ceiling heights of each room (4.1 m for Room #1 and 2.4 m for Room #2).

**Table 3.2** The coordinates of the 6 image sources used to account for reflections of CO from 6 walls located at $x = x_{wall1}, x = x_{wall2}, y = y_{wall1}, y = y_{wall2}, z = z_{wall1}, and z = z_{wall2}$ of a rectangular room with a CO point source positioned at $(x_o, y_o, z_o)$.

<table>
<thead>
<tr>
<th>Image</th>
<th>x-coordinate</th>
<th>y-coordinate</th>
<th>z-coordinate</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$2x_{wall1}$</td>
<td>$y_o$</td>
<td>$z_o$</td>
</tr>
<tr>
<td>2</td>
<td>$2x_{wall2}$</td>
<td>$y_o$</td>
<td>$z_o$</td>
</tr>
<tr>
<td>3</td>
<td>$x_o$</td>
<td>$2y_{wall1}$</td>
<td>$z_o$</td>
</tr>
<tr>
<td>4</td>
<td>$x_o$</td>
<td>$2y_{wall2}$</td>
<td>$z_o$</td>
</tr>
<tr>
<td>5</td>
<td>$x_o$</td>
<td>$y_o$</td>
<td>$2z_{wall1}^{(1)}$</td>
</tr>
<tr>
<td>6</td>
<td>$x_o$</td>
<td>$y_o$</td>
<td>$2z_{wall2}^{(2)}$</td>
</tr>
</tbody>
</table>

(1) $z_{wall1}$ is the position of the assumed ceiling plane at the average ceiling height—the mean of the maximum and minimum ceiling heights of the cathedral ceiling in Room #1 or the vaulted ceiling in Room #2.

(2) $z_{wall2}$ is the position of the floor.
To find the single (isotropic) $K$ value that can best represent the observed spatial spread of CO over the 30-min period, a least-squares method is used. It simultaneously considers and equally weights all 30-37 of the 30-min monitor averages for a given experiment. The Error ($E$) minimized is the sum of the squared difference between each of the measured 30-min time-averaged concentrations, $\bar{C}_{\text{obs}}(x_i, y_i, z_i)$, and the corresponding model estimate—the integration of $C_{\text{model}}(x_i, y_i, z_i, t, K)$, over the monitoring duration, $T$ (30 min), divided by $T$.

$$E(K) = \sum_{i=1}^{N} \left( \bar{C}_{\text{obs}}(x_i, y_i, z_i) - \frac{1}{T} \int_0^T C_{\text{model}}(x_i, y_i, z_i, t, K) dt \right)^2$$

(4)

For eq 4, $N$ is the number of monitors. The integration of $C_{\text{model}}(x_i, y_i, z_i, t, K)$ was numerically approximated using the MATLAB quadrature function ($\text{quadv}$) with a termination tolerance of $10^{-6}$ (Palm, 2005a). $K$ was estimated based on the minimization of eq 4, using the MATLAB nonlinear optimization function ($\text{fminsearch}$) with a termination tolerance on $K$ of $10^{-4}$ (Palm, 2005b). After each computation, we compared the value of the optimized $K$ with that determined using 6 additional image sources (the second nearest image sources) to ensure that a reasonable convergence of the $K$ estimate had been achieved.

In contrast to the pulse release method (Baughman et al., 1994; Drescher et al., 1995, and Klepeis, 1999), this model fitting approach was chosen for use with our continuous CO tracer emissions, which allows us to estimate $K$ directly by fitting spatial CO concentration spreads in the 2 rooms.
3.3 RESULTS AND DISCUSSION

3.3.1 Air Change Rate (ACH)
Table 3.3 summarizes the ACHs and the corresponding $R^2$ from the 2 SF$_6$ monitors (monitors A and B) for the 11 experiments conducted during fall, 2008. With the exception of the 11-4-08 and 11-7-08 (night) experiments, each pair of ACH estimates from the 2 monitors were comparable to each other and each had a regression $R^2$ greater than ~ 0.90. In the 11-7-08 (night) experiment, the ACH estimate of Monitor B was >2 times as large as that of Monitor A. This was the only experiment in which there was one widely opened window (window #2) in the sampling room (see Figure. 3.1), near Monitor B (~2 m from the 8-inch opened window) – in retrospect, this open window is likely to be the reason why Monitor B measured a regional ACH much higher than Monitor A, which was located on the other side of the room. For this experiment, the ACH estimate of Monitor A ($ACH = 0.59 \text{ h}^{-1}$) was used for subsequent analyses.

In general, as the total area of window opening in the house increased (from state 0 to state 3 or 4), the mean air change rate (ACH) of each room increased, ranging from 0.17 to 1.25 h$^{-1}$ for Room #1 and from 0.19 to 5.4 h$^{-1}$ for Room #2. A previous study (Howard-Reed et al., 2002) examined the effect of opening windows on air change rates in 2 occupied residences, one of which was House #1, and found ACH ranging from 0.1 to 3.4 h$^{-1}$. Our results are comparable to these estimates except for the 11-7-08 (morning) experiment where we opened 3 windows as wide as possible in this house (open 16-inches), resulting in an ACH of 5.4 h$^{-1}$.

Compared with Room #1, Room #2 showed greater variation of ACH in different experiments. This variation could be due to its relatively small indoor volume and the more direct ventilation settings (opening windows located in the sampling room). In each room, we conducted 2 experiments with the same window setting. In Room #1, the ACH of the 2 experiments (9-3-08 versus 9-8-08) were comparable to each other (0.57 versus 0.51 h$^{-1}$). However, the ACH of the 2 experiments (11-6-08 (morning) versus 11-6-08 (night)) in Room #2 were not comparable (2.1 versus 0.4 h$^{-1}$). This difference could be
due in part to the diurnal variation of outdoor conditions: the average outdoor wind speed during the daytime experiment (0.9 m s$^{-1}$) was 1.5 times as high as that during the nighttime experiment (0.6 m s$^{-1}$).

Table 3.3 Air change rate (ACH) estimates and the corresponding $R^2$ from the 2 SF$_6$ monitors (monitor A and B) for different natural ventilation settings (state 0- state 3 or 4) of the 11 experiments conducted in the 2 studied rooms.

<table>
<thead>
<tr>
<th>Date</th>
<th>30-min study period</th>
<th>Natural Ventilation Setting</th>
<th>Air Change Rate$^{(3)}$ (h$^{-1}$)</th>
<th>Mean ACH (Error)$^{(4)}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Monitor A ACH ($R^2$)$^{(2)}$</td>
<td>Monitor B ACH ($R^2$)$^{(2)}$</td>
</tr>
<tr>
<td>Room #1</td>
<td></td>
<td></td>
<td>Monitor A ACH ($R^2$)$^{(2)}$</td>
<td>Monitor B ACH ($R^2$)$^{(2)}$</td>
</tr>
<tr>
<td>9-2-08</td>
<td>afternoon</td>
<td>State 0</td>
<td>0.16 (0.972)</td>
<td>0.18 (0.984)</td>
</tr>
<tr>
<td>9-3-08</td>
<td>morning</td>
<td>State 1</td>
<td>0.58 (0.995)</td>
<td>0.55 (0.997)</td>
</tr>
<tr>
<td>9-8-08</td>
<td>morning</td>
<td>State 1</td>
<td>0.50 (0.995)</td>
<td>0.51 (0.993)</td>
</tr>
<tr>
<td>9-4-08</td>
<td>morning</td>
<td>State 2</td>
<td>0.81 (0.992)</td>
<td>0.75 (0.990)</td>
</tr>
<tr>
<td>9-6-08</td>
<td>morning</td>
<td>State 3</td>
<td>1.26 (0.987)</td>
<td>1.24 (0.992)</td>
</tr>
<tr>
<td>Room #2</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>11-4-08</td>
<td>afternoon</td>
<td>State 0</td>
<td>0.37(0.993)</td>
<td>malfunction</td>
</tr>
<tr>
<td>11-5-08</td>
<td>morning</td>
<td>State 1</td>
<td>0.17(0.897)</td>
<td>0.21(0.957)</td>
</tr>
<tr>
<td>11-7-08</td>
<td>night</td>
<td>State 2</td>
<td>0.59(0.930)</td>
<td>1.38(0.990)</td>
</tr>
<tr>
<td>11-6-08</td>
<td>morning</td>
<td>State 3</td>
<td>1.95 (0.984)</td>
<td>2.20(0.989)</td>
</tr>
<tr>
<td>11-6-08</td>
<td>night</td>
<td>State 3</td>
<td>0.37(0.975)</td>
<td>0.44(0.957)</td>
</tr>
<tr>
<td>11-7-08</td>
<td>morning</td>
<td>State 4</td>
<td>5.40(0.986)</td>
<td>5.40(0.996)</td>
</tr>
</tbody>
</table>

$^{(1)}$Air change rates (ACH) were estimated from the slope of the log-linear regression line between SF$_6$ concentration and time.

$^{(2)}$ $R^2$ value of the log-linear regression between SF$_6$ concentration and time.

$^{(3)}$ Excludes ACH estimate of Monitor B.

$^{(4)}$Error = 100% × |ACH of monitor A – ACH of monitor B| / Mean ACH
3.3.2 Turbulent Diffusion Coefficient ($K$)

Using the real-time CO monitoring array measurements in each experiment, we computed the 30-min time-averaged concentrations for all monitored indoor positions. Figure 3.2(a) and 3.2(b) show examples of the typical time-averaged concentration distributions on the measured x-y plane within 2 m from the source at the origin, plotted using the 2-D interpolation function (griddata,'v4') in MATLAB (Trauth, 2007). As seen in these 2 plots, the CO distributions were in general symmetrical with respect to the source in the 2 rooms. These typical plots support the assumption/simplification that under natural ventilation conditions, the magnitude of mean/time-averaged advection is negligible compared to turbulent diffusion indoors. On the other hand, the concentration distribution was less symmetrical (Figure 3.2(c)) for the one experimental period when a pronounced discrepancy between the 2 $ACH$ estimates was seen. This may be due to the open window in the room causing a more pronounced preferential directional air flow and/or spatially non-uniform mixing ($K$ varying with indoor locations) during this 30-min experimental period.
Figure 3.2. Three examples of the spatial distributions of 30-min time-averaged CO concentration on the measured x-y plane within 2 m from the continuous CO tracer source at the origin.
Given the time-averaged CO measurements in space of each experiment, we used eq 3 and the measured mass emission rate of the CO source to find the optimal isotropic turbulent coefficient ($K$). Table 3.4 summarizes estimates of $K$ from the 11 experiments in the 2 rooms. All $K$ values found were consistent with those determined by adding 6 additional image sources with overall mean absolute error of 5.8 ppm between measured and modeled 30-min time-averaged concentrations at all monitored indoor positions for 30-min measurements of 0.3-105.3 ppm Compared with Room #1 ($K = 0.002-0.007 \text{ m}^2\text{s}^{-1}$), Room #2 showed greater variation in $K$, ranging from 0.003 to 0.015 m$^2$s$^{-1}$.

### Table 3.4 Turbulent diffusion coefficient estimates ($K$) from the 11 experiments conducted in the 2 studied rooms at different air change rates ($ACH$).

<table>
<thead>
<tr>
<th>Study period</th>
<th>Room #1</th>
<th></th>
<th>Room #2</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>ACH ($\text{h}^{-1}$)</td>
<td>9-2-08</td>
<td>9-8-08</td>
<td>9-3-08</td>
<td>9-4-08</td>
</tr>
<tr>
<td>$K$ ($\text{m}^2\text{s}^{-1}$)</td>
<td>0.00190</td>
<td>0.00407</td>
<td>0.00459</td>
<td>0.00528</td>
</tr>
<tr>
<td>Subplot</td>
<td>a</td>
<td>b</td>
<td>c</td>
<td>d</td>
</tr>
<tr>
<td>$R^2$</td>
<td>0.969</td>
<td>0.963</td>
<td>0.985</td>
<td>0.942</td>
</tr>
<tr>
<td>$R^2$</td>
<td>0.961</td>
<td>0.977</td>
<td>0.994</td>
<td>0.953</td>
</tr>
<tr>
<td>Mean outdoor wind speed ($\text{m s}^{-1}$)</td>
<td>0.34</td>
<td>0.60</td>
<td>0.42</td>
<td>0.43</td>
</tr>
</tbody>
</table>

1. Corresponding subplot in Figure 3.3 for each experiment, comparing the modeled with measured dimensionless CO concentration ($C/C_o$) at different distances from the source.
2. Slope of the linear regression line between modeled and measured 30-min time- and radially averaged CO concentrations at different distances from the source.
3. $R^2$ value of the linear regression between modeled and measured 30-min time- and radially averaged CO concentrations at different distances from the source.
4. One CO monitor at 4 m from the source malfunctioned, so $K$ was determined using the rest of 36 CO monitor measurements.
5. Excludes 4 measurements at 0.25 m from the source.
Our estimates collected in two residential indoor spaces are at the lower end of the range of reported $K$ values (0.001-0.2 m$^2$/s) measured in occupational settings, such as indoor industrial environments (Wadden et al., 1989, Scheff et al, 1992, Conroy et al, 1995, Demou et al, 2009). One possibility is that our natural/unforced ventilation settings in a home introduced less air mixing than mechanical ventilation in occupational workplaces, reducing the magnitude of turbulence. Also, in the absence of mechanical air mixing, vertical indoor thermal stratification is more likely, further attenuating the dispersion of air pollutants indoors. Another possible consideration involves differences in the experimental setup: our array of 30-37 monitors was deployed over the entire indoor space, providing spatially well-averaged estimates of $K$. These average values could be quite different from results involving a few monitors, one or a few axes, and/or shorter averaging times.

Using the 30-min monitoring array datasets, we examined $K$ estimated from a selected single-direction measurements (along the positive x-direction), and from a shorter averaging time (10 min). The resulting $K$ values showed much more variation, covering 2 orders of magnitude. This result implies there is greater uncertainty in estimated $K$ when intensive spatial and temporal measurements are not feasible. It also suggests the difficulty to model deterministically exposures over short time periods (e.g., ~10 min or less) at a specific position. This uncertainty likely arises because of transient directionality in the emitted plume due to turbulent mixing indoors.

### 3.3.3 Relationship between $ACH$ and $K$

To examine how the measured spatial spreads of CO differed from each other for different air change rates, and how well the isotropic eddy diffusion model can describe each of the measured CO concentrations as a function of distance, we radially averaged (across all monitors at each radial distance) the computed 30-min time-averaged measurements. These results were compared with the concentrations at each radial distance modeled by eq 3, using the optimized $K$ value (see Table 3.4) for each of the 11 experiments. Both measured and modeled concentrations ($C$) were then normalized by a
reference concentration \( (C_o) \) — the 30-min time-averaged concentration predicted by the well-mixed mass balance model:

\[
C_o = \frac{1}{T} \int_0^T \frac{q}{ACH \cdot V} \left[ 1 - \exp(-ACH \cdot t) \right] dt
\]  

(5)

For eq 5, \( T \) is the averaging time (1800 s), \( q \) (\( \mu g/s \)) is the CO mass emission rate, \( ACH \) (s\(^{-1}\)) is the air change rate, and \( V \) (m\(^3\)) is the volume of the indoor space. This approach normalized for variations in the CO emission rate across different experiments and reflected how concentrations at different radial distances from the source compare with the predictions of the well-mixed mass balance model.

Figure 3.3(a)-(k) plots the comparison between measured and modeled dimensionless concentrations \( (C/C_o) \) for the 5 experiments in Room #1 and the 6 experiments in Room #2. The subplots for each room are stacked with \( ACH \) increasing from top to bottom. The different scales of \( C/C_o \) between Room #1 and Room #2 was due to normalizing by \( C_o \): the emission rates, \( q \) used in the 2 rooms were comparable, but the volume of Room #1 is \(~2.7\) times as large as Room #2.
Figure 3.3(a)-(k). Comparison between measured and modeled dimensionless CO concentrations ($C/C_o$) for the 5 experiments in Room #1 and 6 experiments in Room #2. The subplots (small graphs) for each room are stacked with $ACH$ increasing from top to bottom. $C$ is the initial 30-min time- and radially averaged CO concentration, whereas $C_o$ is the initial 30-min time-averaged concentration predicted by the well-mixed mass balance model (see eq 5).
For each experiment, as the distances from the CO source decrease, the measured $C/C_0$ ratio increases noticeably – it is below 1 farthest away from the source, but above 1 for radial distances $< 1$ m. This reflects the non-instantaneous mixing of indoor air during the active emission period. In general, as air change rate increases, CO becomes more rapidly distributed over the indoor space, resulting in a lower $C/C_0$ in close proximity to the source. This trend is especially noticeable for the 5 experiments in Room #1(Figure 3.3(a)-3.3(e)).

Comparing the modeled to measured $C/C_0$, we found that for most of the experiments, the isotropic eddy diffusion model (eq 2) can describe the observed averaged CO profiles with the radial distances without significant error. The one exception (Figure 3.3(f)), had lower $C/C_0$ at 0.25 m than at 0.5 m from the source. This unusual measured concentration profile could have been associated with a time of sustained directional air motion near the source, making the 0.25 m monitors placed at 4 angles unable to fully capture the expected higher concentration near the source.

To examine quantitatively how well each $K$ value can represent the observed concentration distribution in space, we performed the linear regression between modeled and measured 30-min time- and radially averaged concentrations at different distances from the source for each experiment and examined the resulting slope ($m$) and $R^2$ value of the regression line (see Table 3.4). Compared with Room #2 that had $m = 0.714$-$0.998$ and $R^2 = 0.537$-$0.984$, Room #1 showed more consistently satisfactory fitting results with $m = 0.942$-$0.985$ and $R^2 = 0.953$-$0.994$. This difference could be due to the larger indoor volume of Room #1 and its indirect ventilation settings (opening the windows in other rooms of House #1), making it less susceptible to directional air flow introduced from windows. Among all 11 experiments, the experiment on 11-5-08 had the least satisfactory fitting result ($m = 0.71$ and $R^2 = 0.54$) due to its underestimated measurement at 0.25 m from the source. To better represent the turbulent diffusion coefficient for this experiment, we carried out the least-squares optimization without the 4 0.25-m measurements ($N = 26$). This approach gave a $K$ value of $0.00107$ m$^2$ s$^{-1}$ ($m = 0.77$; $R^2 = 0.71$), which was
used in place of the original value (0.00260 m² s⁻¹) for the subsequent analysis (see Table 3.4).

As noted earlier, eq 3 does not account for the removal of CO via air exchange. The 2 experiments performed at the highest ACH (> 2 h⁻¹) had fitting results (m > 0.95 and R² > 0.88) consistent with those at lower ACH values, indicating that neglecting the removal of CO due to air exchange over the short-term experimental duration (30 min) did not cause noticeable detriments in the regressions at higher ACH values.

Figure 3.4 shows the relationship between turbulent diffusion coefficients (K) and the air change rates (ACH) for the 2 indoor spaces. In general, as the ACH increased, the magnitude of the turbulent diffusion coefficient increased correspondingly. The trend observed is steeper in Room #1 than in Room #2. In Room #2, one K estimate at the ACH of ~ 0.5 h⁻¹ was noticeably higher than the other 2 measures and deviated from the general trend. Because this was the only K in Room #2 measured during an (sunny and clear) early afternoon, we hypothesize that this result could be associated with the stronger thermally induced mixing due to incoming solar radiation, further contributing the magnitude of turbulent diffusion indoors. If the thermal energy input is relatively strong while the ACH is very low, the effect of thermal mixing could become important (Baughman et al, 1994).

The correlation of K for each room with the corresponding ACH is significant (p < 0.001), with R² of 0.97 and 0.95 for Room #1 and Room #2, respectively. However, the slope of the regression line for Room #1 (m = 0.004) was twice as large as that for Room #2 (m = 0.002). This variation could be due to the significant difference in indoor volume between the two rooms studied (Room #1 is ~2.7 times as large as Room #2) – while ACH factors in the room dimensions, K does not.

To generalize more fully the data sets collected in the two rooms, we defined the characteristic length scale of the room (L) as the cube root of the indoor volume (5.41 m for Room #1 and 3.90 m for Room #2). By dividing K with L², we normalized K for the
variation in indoor volume between the 2 rooms studied. This normalization of $K$ has the same units of inverse time as $ACH$. Here, $K / L^2$ can be thought of as the indoor air mixing rate — the reciprocal of the time scale of turbulent mixing indoors. This formulation of the turbulent mixing time scale has been used in many water mass transfer applications (e.g., Fischer et al., 1979).

Figure 3.4. Associations between turbulent diffusion coefficients ($K$) and the air change rates ($ACH$) of the 2 rooms studied (Room #1 and Room #2).

Figure 3.5 plots the relationship between the air mixing rate ($K / L^2$) and air change rate ($ACH$) for the two rooms, combined. As shown, the measurements collected from the two different indoor spaces closely align, resulting in a significant overall linear correlation ($p < 0.001, n = 11$) with $R^2$ of 0.94. This is consistent with the theoretical expectation that
the rate of CO loss via turbulent diffusion through opened windows is equivalent to the volume-normalized outflow rate of indoor air—$ACH$, based on the mass balance and scaling derivations. The positive $y$-intercept of the linear regression (0.27) could be associated with the thermal energy input (i.e. sunlight heating on wall surfaces) in the room further contributing to the magnitude of air mixing indoors.

The 95% confidence interval (dash lines) shows that the uncertainty in predicted air mixing rate decreases with smaller $ACH$, due to more available data points. For 95% of U.S. residences that have $ACH < 2$ h$^{-1}$ (Wilson et al., 1996; Pandian et al., 1998), the corresponding confidence intervals are less than ±0.2 h$^{-1}$. Although the vertical temperature gradients of Room #1 (0.7-1.3 °F m$^{-1}$) were ~7 times as large as those of Room #2 (0.1-0.2 °F m$^{-1}$), no pronounced deviations between the air mixing rate estimates from the 2 indoor spaces were observed within the range of temperature gradients seen in our 11 experiments (0.1-1.3 °F m$^{-1}$).

Using the fitted lognormal distribution ($\mu = -0.64; \sigma = 0.82$) for $ACH$ in 2844 U.S. residences (Murray and Burmaster, 1995), we applied the empirical relation between $K / L^2$ and $ACH$ (Figure 3.5) to estimate $K$ for a typical small bedroom and a large living/family room with assumed volumes of 25 and 150 m$^3$ ($L = 2.9$ and 5.3 m), respectively. The resulting distribution of $K$ for the bedroom gives a mean, first, second (median), and third quartiles of 0.0017, 0.0011, 0.0014, and 0.0019 m$^2$/s, respectively compared to 0.0056, 0.0035, 0.0046, and 0.0065 for the living/family room.

Previous studies examined the mixing of a pulse release in an experimental room (Baughman et al, 1994 and Drescher et al, 1995) and in a residential bedroom and a tavern (Klepeis, 1999). They found indoor mixing times ranging from 2 to 42 min, based on applying an empirical criterion of CV<10% to the measurements. Our mixing time scale estimates (17-260 min) should not be directly compared with these experimental values, because they are based on a dimensional analysis approach, combining a turbulence measure ($K$) with a characteristic length scale ($L$) to obtain a turbulent mixing time scale.
The observed significant linear correlation between the air mixing rate \( (K / L^2) \) and the air change rate \( (ACH) \) suggests the possibility of predicting the turbulent diffusion coefficient \( (K) \) using just parameters that can be readily measured in a typical indoor field study: the \( ACH \) and the room dimensions. We expect that the modeled \( K \) values could theoretically be used along with appropriate eddy diffusion models to estimate proximity exposures in the presence of either an instantaneous or a continuous indoor air pollution point source.

**Figure 3.5.** Relationship between the air mixing rate \( (K / L^2) \) and air change rate \( (ACH) \) of the 2 indoor spaces studied (Room #1 and Room #2). The air mixing rate was represented as the turbulent diffusion coefficient \( (K) \) normalized by the square of the length scale of room dimensions \( (L) \), which was calculated as the cube root of the indoor volume.

\[
K / L^2 = 0.61 \, ACH + 0.27;
\]

\( (R^2 = 0.94, \, n = 11) \)
3.4 SUMMARY AND IMPLICATIONS

This study examined the relationship between indoor turbulent diffusion coefficients and air change rates in 2 naturally ventilated residential rooms in 2 different homes for a range of ventilation conditions. Our results showed that:

- Under natural ventilation conditions where advective air flow was not significant, the isotropic eddy diffusion formulation can describe the measured 30-min time- and radially averaged concentration profiles at breathing height (1 m from the floor) with reasonable accuracy.

- In 2 naturally ventilated indoor spaces, we found that the magnitudes of the turbulent diffusion coefficient ranged from 0.001 to 0.015 m$^2$s$^{-1}$ for air change rates of 0.2-5.4 h$^{-1}$.

- In each room studied, we found that as the air change rate increased, the magnitude of the turbulent diffusion coefficient increased correspondingly, resulting in a significant positive linear correlation ($p < 0.001$) with $R^2 > 0.94$.

- Representing the air mixing rate as the turbulent diffusion coefficient divided by the square of the indoor volume length scale, we were able to normalize for variations in room size and found a significant overall positive linear correlation between the air change rates and the air mixing rates for the two rooms combined ($p < 0.001$, $n = 11$) with $R^2 = 0.94$.

Although widely assumed in indoor proximity exposure models found in occupational literature, the concept of isotropic eddy diffusion has not been rigorously tested previously due to an insufficient number of real-time monitoring results, and it has never before been applied to a home. The reasonable agreement between modeled and measured average concentrations in our residential monitoring array experiments demonstrates that currently available indoor turbulent diffusion models can predict exposure in close proximity to indoor sources within naturally ventilated spaces, over an averaging time scale of 30 min.
Estimating a turbulent diffusion coefficient that can represent the overall mixing characteristics of the entire indoor space requires simultaneous measurements of a large number of monitors deployed at different indoor positions. This massive monitor array is not feasible for most indoor air quality investigators. The significant linear correlation between indoor air mixing rates and air change rates observed in this study suggests a possible new approach for predicting the turbulent coefficient for an indoor space of interest, using just the air change rate and the dimensions of the indoor space.

A number of studies have shown that higher exposures occur near an actively emitting indoor air pollutant source (Rodes, et al, 1991, Furtaw et al., 1996; McBride et al., 1999, McBride 2002; Ferro et al., 1999, 2004, 2009; Acevedo-Bolton et al., 2010). The proximity effect is relevant to many common indoor activities such as smoking, cooking, and household cleaning. Previously no physics-based characterizations of the exposure proximity effect have been made in real residential environments. The proximity effect and its relationship to turbulent diffusion indoors is complex, but we believe this model has made significant progress toward understanding the factors that affect the proximity effect indoors.
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REFERENCES


CHAPTER 4

Modeling the Effect of Proximity on Exposure to an Indoor Active Air Pollution Source in Naturally Ventilated Rooms: An Application of the Stochastic Random Walk Process


ABSTRACT

A three-dimensional random-walk model has been developed to model the higher exposure in close proximity to an active air pollution point source in naturally ventilated indoor spaces. The model incorporates physical processes of anisotropic turbulent diffusion, removal of the air pollutant, and air pollutant reflection from wall boundaries. To model the highly variable exposure near an active source, we developed a new piecewise random walk algorithm to stochastically simulate transient directional air motions of turbulent mixing indoors. The distribution of different exposure cases generated using this model reasonably covered the range of experimental measurements collected in 2 houses, while preserving ensemble averages satisfying the principle of Fickian diffusion. The presented modeling concept offers a new starting point for predicting transient peak exposures close to an active source under turbulent mixing conditions, with potential applications not only involving indoors but also other environmental locations.
4.1 INTRODUCTION

4.1.1 Source Proximity Effect on Personal Exposure

Populations in developed nations spend most of the time in indoor environments, so personal exposure to indoor air pollution constitutes a significant fraction of total exposure, and becomes an important consideration for health risk assessment (Robinson et al., 1991; Smith, 1993; Nelson et al., 1994). Typically, exposure to an indoor emission source has been modeled by the well-mixed mass balance model (e.g. Shair and Heitner, 1974; Hayes, 1991; Keil, 1998; von Grote et al, 2003; Vernez et al, 2006), which assumes that air pollutants emitted indoors become instantaneously, completely well-mixed. Therefore, the concentrations are represented as homogeneous throughout an indoor space, but varying with time due to the emissions and removals of the pollutants. Since the transient imperfect mixing period immediately after the release is typically less than 1 h in indoor environments (Baughman et al, 1994; Drescher et al, 1995; Klepeis, 1999), this model can provide a simple and accurate method to approximate the long-term exposure to short-duration indoor emissions when the source emission and mixing time scales are much smaller than the duration over which the time-averaged concentration (the estimate of exposure) is considered. However, for a continuous source releasing air pollutants over a duration that is comparable to the exposure time of interest, the imperfect mixing during the emission period becomes important to consider. During this active source period, exposures in close proximity to the source are expected to be substantially higher than those further away from it – this source “proximity effect” cannot be captured by the uniform mixing model commonly used in indoor exposure studies.

This source proximity effect is one reason why indoor measurements using personal monitors worn by people have typically been higher than those using stationary monitors placed at fixed indoor locations (Rodes et al., 1991; Özkaynak et al., 1996). For example, exposures to particulate matter (PM) for a person performing indoor activities that resuspend particles were up to 8.5 times as high as those measured by the stationary
monitor placed 10-m away from the person during the activity and emission periods (Ferro et al., 1999, 2004).

To examine the indoor source proximity effect, controlled experiments using multiple real-time monitors have been conducted to capture the spatial and temporal variation of air pollutant concentrations in indoor spaces. Using a tracer gas, Furtaw et al. (1996) found that the exposure at arm's length (approximately 0.4 m) from the source exceeded the theoretical well-mixed concentration by a ratio of about 2:1. McBride et al. (1999, 2002) placed monitors at different distances (0.5-5.4 m) from a continuous CO source, and they found that real-time concentrations within 2 m of the source were up to ~4 times as high as the predictions of the well-mixed mass balance model. Most recently, Acevedo-Bolton et al. (2010) reported even higher elevations in concentration close to the source, along with pronounced fluctuations with time – extremely high concentration peaks lasting for only a few seconds, called microplumes. These results imply that exposures in close proximity to the source are elevated and highly variable.

4.1.2 Indoor Dispersion Modeling

4.1.2.1 Deterministic model

2-compartment model. An early attempt to model the higher exposure near an active source utilized a 2-compartment model. In this model, the indoor space was conceptually divided into 2 well-mixed zones—the near-field (NF) zone containing the emission source, and the far-field (FF) zone representing the rest of the room. Air pollutants were transferred between these 2 well-mixed zones to model the pollutant dispersion into the indoor air (Furtaw et al., 1996; Nicas, 1996, 2000). Although this type of modeling approach can capture the elevated exposure in close proximity to the source, it inevitably yields a discontinuity in concentration at the boundary of the two zones (Nicas, 2001). Also, it requires additional experimental air flow parameters (i.e. air velocity) to describe the interzonal air exchange (Bennett at al., 2003; Demou et al., 2009).

Turbulent diffusion model. To better model the dispersion of air pollutants from an active indoor source, several models have been proposed and used, typically for
occupational and industrial applications. These models assume that for an indoor space enclosed by walls, there is no persistent directional drift. However, there is random motion of air (zero velocity on average) which allows air pollutants to be dispersed in a symmetrical manner, described as a turbulent diffusion process (Wadden et al., 1989; Conroy et al., 1995; Drivas et al., 1996; Keil et al., 1997; Nicas, 2001). By assuming that advection is negligible compared with turbulent diffusion, analytical solutions to Fick’s Second Law of Diffusion can be utilized to characterize the indoor proximity effect.

Assuming an isotropic turbulent diffusion condition for a continuous indoor point source, one can use eq 1, containing an error function, to describe the concentration distribution over time and space (Crank, 1975):

\[
C = \frac{q}{4\pi K r} \left(1 - \text{erf} \left( \frac{r}{\sqrt{4Kr}} \right) \right)
\]  

(1)

For eq 1, \(C (\mu g \text{ m}^{-3})\) is the air pollutant concentration; \(q (\mu g \text{ s}^{-1})\) is the mass emission rate of the air pollutant; \(K (\text{m}^2 \text{ s}^{-1})\) is the isotropic eddy diffusion coefficient for the indoor space; \(r (\text{m})\) is the distance from the indoor point source; and \(t (\text{s})\) is time. To account for the air pollutant reflection from the floor, Wadden et al. (1989), Conroy et al. (1995), and Keil et al. (1997) modeled concentration as a function of the distance from the source by multiplying eq 1 by a factor of 2. Although the great simplicity of this model (eq 1) allows concentration to be readily calculated, overestimation of exposure levels is expected, because the equation fails to account for the removal of indoor air pollutants via air exchange or surface deposition/adsorption. This error can become large as the exposure time scales of interest increase.

To incorporate the removal of indoor air pollutants, the solution (eq 2) for a pulse release of the air pollutant can be considered (Crank, 1975):

\[
C = \frac{m}{(4\pi K t)^{1.5}} \exp \left( \frac{-r^2}{4Kt} \right)
\]  

(2)
For eq 2, \( C (\mu g m^{-3}) \) is the air pollutant concentration; \( m (\mu g) \) is the total air pollutant mass emitted from the point source; \( K (m^2 s^{-1}) \) is the isotropic eddy diffusion coefficient for the indoor space; \( r (m) \) is the distance from the indoor point source; and \( t (s) \) is the elapsed time since the pulse release. Drivas et al. (1996) has multiplied eq 2 by an exponential removal term to include the first-order removal of air pollutants for an instantaneous indoor emission source.

To include removal processes for a continuous source, one can treat the continuous emissions as a sequence of pulse releases. By superposing the solutions (with the exponential removal term) for the sequential pulse releases, one can predict the spatial concentration distribution in the presence of a continuous source while preserving the effect of indoor pollutant removal processes. The drawback of this approach is that the superposition of the solutions involves numerically integrating eq 2 with respect to time.

For an indoor environment, it is important to account for the interaction between wall surfaces and air pollutants, especially for the case where an indoor source is located in close proximity to the wall boundaries (i.e. the floor). To accomplish this analytically, one can introduce “image sources” to enforce either total reflection or total adsorption boundary conditions at wall positions. However, for an indoor space enclosed by 6 walls, a large series of image sources may need to be summed to ensure a convergence of the solution. Using eq 2 in a Cartesian coordinate system, Drivas et al. (1996) have employed this method to model the pollutant reflection from indoor wall surfaces.

**Computational fluid dynamics (CFD).** When forced air flow is introduced into an indoor space (i.e. mechanical ventilation), advection of air pollutants becomes important to consider. In this case, a computational fluid dynamics (CFD) approach is commonly used to model the dispersion of air pollutants (e.g. Andersson and Alenius, 1996; Buchanan and Dunn-Rankin, 1998; Gadgil et al., 2003; Beghein et al., 2005; Chang et al., 2006). The mathematical formulation is typically based on conservation of mass and momentum from which the velocity field that governs the distribution of air pollutants can be portrayed (Beghein et al., 2005; Chang et al., 2006). Although more
comprehensive mass transport processes are incorporated, CFD models require additional input parameters not routinely measured in typical indoor air quality (IAQ) investigations (i.e. supply air velocity; air pressure).

4.1.2.2 Stochastic model
In contrast to these deterministic approaches, Nicas (2001) invoked the theory of random walk to describe eddy diffusion transport indoors, implementing it in a stochastic Markov Chain model. In this model, the room is divided into cubic cells. For each time step, a particle in a given cell can hold its position or move to one of the 6 physically contiguous cells with equal chance. When a large number of particles are released, particles can be spatially distributed in a manner that is similar to the process of the eddy diffusion transport.

The fundamental concept of using the stochastic particle random walk theory offers an alternative approach to resolve the complex indoor mixing process with simplicity. However, to simulate 3-dimensional transport, the computation of particle transport among these cubic cells requires a large amount of computational effort, especially for the case where higher spatial resolution of the pollutant distribution is of interest. In addition, like the currently available analytical indoor turbulent diffusion models, it does not account for the anisotropic mixing that could occur in thermally stratified indoor spaces (Gao et al, 2009).

This study aims to model the proximity effect for personal exposure to an active air pollution point source in naturally ventilated indoor environments (in the absence of mechanical or forced air flow). To predict more accurately the higher exposure close to an active indoor source, our first goal is to construct a 3-dimensional random-walk particle tracking model that can resolve anisotropic diffusion along with pollutant removal and wall reflection indoors, based on input parameters that are routinely measured in IAQ investigations. In an effort to model the greater variability of exposure in close proximity to the source, our second goal is to develop a new random walk algorithm that can stochastically simulate transient directional air movements of turbulent
mixing indoors. As opposed to the Eulerian approach, the Lagrangian modeling platform was chosen to allow (i) more detailed delineations of turbulent mixing in space (i.e. the microscopic structure of turbulent eddies), and (ii) Monte Carlo simulation for the random directionality of turbulence in time.

4.2 MODEL FORMULATION

4.2.1 Modeling the Higher Exposure in Close Proximity to an Active Source

To model the mass transfer of air pollutants from an active point source, we assume that there is no pronounced, sustained advection in naturally ventilated rooms. Thus, the magnitude of mean advective air flow is negligible compared to turbulent diffusion over the exposure time scales of interest (Péclet number$^3$ $<< 1$). We further assume that the turbulent mixing of indoor pollutants is spatially uniform. This simplification allows us to utilize spatially-averaged turbulent diffusion coefficients, which can be determined experimentally, as the model input variables.

In the absence of mechanical ventilation and mixing, indoor thermal stratification can be significant (Webster et al., 2002; Wan and Chao, 2005). This stratification inhibits vertical mixing in a room, making the turbulent diffusion in the vertical direction relatively weaker than in the horizontal direction. To consider this anisotropy, we express the general indoor mass transfer equation as:

$$\frac{\partial C}{\partial t} = K_h \left( \frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} \right) + K_v \frac{\partial^2 C}{\partial z^2} + E - S \tag{3}$$

In eq 3, $C$ ($\mu g m^{-3}$) is the indoor air pollutant concentration; $K_h$ and $K_v$ ($m^2 s^{-1}$) are the turbulent diffusion coefficients in the horizontal and vertical directions, respectively; and $E$ and $S$ ($\mu g m^{-3} s^{-1}$) are the emission and removal rates, respectively. Assuming that the removal of indoor air pollutants due to indoor-outdoor air exchange and surface

$^3$ Péclet number is the ratio of the rate of advection to the rate of diffusion, calculated by velocity times a characteristic length scale divided by diffusion coefficient (e.g., Charbeneau, 2000).
deposition and adsorption follows a first order decrease (Drivas et al., 1996), one can express removal flux, $S$ as:

$$ S = (ACH + k)C $$

(4)

In eq 4, $ACH$ ($s^{-1}$) is the air exchange rate of an indoor space, and $k$ ($s^{-1}$) is the removal rate of air pollutants due to surface deposition and adsorption, both of which are routinely measured in typical IAQ investigations.

We solve eq 3 by invoking a Lagrangian approach, where we introduce a large number of air parcels in a system, and then track their trajectories individually. By interpreting the time-varying distributions of the air parcels in space, we can evaluate exposures at different indoor positions over the active source period. Given the general indoor mass transfer equation (eq 3), we aim to simulate (i) the anisotropic indoor turbulent diffusion; (ii) the continuous point source release; (iii) the first order removal of the air pollutant; and (iv) the air pollutant reflection from wall boundaries within the Lagrangian framework. These simulations are discussed in the next four subsections, respectively.

**Anisotropic eddy diffusion.** To simulate the eddy diffusion of an air pollutant in an indoor environment, we invoke the stochastic random walk theory where the diffusive displacements of air parcels are quantified for each sequential time step ($\Delta t$) as mutually independent random variables from a common distribution (typically a normal distribution) with mean $\mu = 0$ and variance $\sigma^2 = 2K\Delta t$. The probabilistic time-step displacements allow us to employ the particle tracking algorithm to simulate random movements of parcels and ultimately solve the diffusion transport equation (Kitanidis, 1994; James and Chrysikopoulos, 2001). For anisotropic transport in the 3-dimensional Cartesian system, the algorithms can be formulated as:

---

4 In the simulation, each air parcel is a singular point in space (with infinitely small volume) carrying the mass of the air pollutant from one position to another at each time step based on the mass transfer principles.
\[ X^n = X^{n-1} + (2K_n \Delta t)^{1/2} \alpha, \quad n = 1, 2, \ldots \] \hspace{2cm} (5a)

\[ Y^n = Y^{n-1} + (2K_n \Delta t)^{1/2} \beta, \quad n = 1, 2, \ldots \] \hspace{2cm} (5b)

\[ Z^n = Z^{n-1} + (2K_n \Delta t)^{1/2} \gamma, \quad n = 1, 2, \ldots \] \hspace{2cm} (5c)

In eq 5(a)-5(c), \( X^n \), \( Y^n \), and \( Z^n \) are arrays of the same size representing the 3-D positions of air parcels at the \( nth \) time step. \( X^{n-1} \), \( Y^{n-1} \), and \( Z^{n-1} \) are the position arrays at the previous (n-1)th time step. \( \alpha \), \( \beta \), and \( \gamma \) are arrays (with the same size as the position arrays) containing mutually independent random variables from a unit normal distribution, \( N(\mu = 0, \sigma^2 = 1) \). This array algorithm allows simultaneous computations for all air parcels at each time step. For example, for the computations of 4 air parcels, a total of 12 random numbers are generated from the unit normal distribution at the same time to model the displacements of 4 air parcels in the \( x \)-, \( y \)-, and \( z \)-directions based on the corresponding \( K \) for each decomposed direction.

**Continuous source.** To simulate an active emission source with a constant emission rate, we introduce the same number of air parcels for each time step at the defined source position to create a sequence of pulse releases at the source release point. To accomplish this, the same number of source positions is added to the position array for each time step, allowing newly released air parcels to be transported/updated along with the existing air parcels in the same manner.

**Pollutant removal.** Using the experimental parameters of \( ACH \) and \( k \), we formulate the removal of pollutants based on the time evolution of air parcels in an indoor space. For a continuous source, the removal of air pollutants is simulated by tracking the life spans of the sequentially released puffs of air parcels, in order to consider their elapsed times for removal individually. Using a parallel array (with the same size as the position arrays), we further define that each of the released air parcels contains the same initial particle
number \((N_o)\). After each time step, each parcel of particles\(^5\) is subjected to a first-order removal. For a parcel of particles that is released \(m\) time steps before the \(n\)th time step, the residual particle number in the parcel can be computed as:

\[
N^n = N_0^{n-m} \exp(- (ACH + k)m\Delta t)
\]

In eq 6, \(N^n\) is the residual particle number of a parcel (released at \((n-m)\)th time step) at the \(n\)th time step. \(N_0^{n-m}\) is the initial particle number of the parcel. For a continuous source, the total amount of particles in a compartmental space of interest at a certain time is the superposition of the numbers of residual particles of different life spans.

**Wall reflection.** The first-order removal algorithm already accounts for the losses associated with indoor-outdoor air exchange and surface deposition and adsorption. Thus, a total reflection boundary condition is needed for the interaction of air parcels with the walls to maintain mass balance. This is achieved by an additional algorithm that corrects the positions of those air parcels the model has transported by diffusion across the wall boundaries. Analogous to the analytical approach of introducing source mirror images, we invoke geometric algebra to calculate the positions of air parcels that are reflecting from the 6 walls of a rectangular room. For instance, the correction algorithm for the \(x\)-coordinates of air parcels for a wall plane positioned at \(x_b\) is formulated as:

\[
x = 2x_b - x^*
\]

In eq 7, \(x^*\) represents the initial positions of air parcels that have been mistakenly transported beyond the wall boundaries, whereas \(x\) are the corrected air parcel positions after wall refection. This algorithm (eq 7) is based on the geometric algebra for 2 points (\(x\) and \(x^*\)) symmetric with respect to the plane at \(x = x_b\). This correction formulation is also applied to \(y\)- and \(z\)-coordinates of air parcels at the same time step of computation.

---

\(^5\) In the simulation, particles within each moving air parcel are given the same position in space, but the number of particles decreases with time based on eq 6.
Concentration interpretation. In the Lagrangian particle tracking method, concentration has been interpreted by counting the number of air parcels within a compartmental space of interest, with each air parcel given the same mass. To account for the pollutant removal, we instead count the total number of residual particles within the compartment to evaluate the bulk (space-averaged) mass concentration at a specific time.

In this model, an arbitrary particle number emission rate (number of particles introduced per time step) is first defined. By providing the actual mass emission rate of an indoor source, the bulk mass concentration at a compartmental space can be calculated as:

$$C(x_c, y_c, z_c, t) = \frac{\frac{\dot{N}(x_c, y_c, z_c, t)}{V}}{\frac{\dot{M}}{S}}$$

(8)

In eq 8, $C(x_c, y_c, z_c, t)$ ($\mu g \ m^{-3}$) is the bulk mass concentration for a compartmental space centered at $(x_c, y_c, z_c)$ at time $t$ (s). $N(x_c, y_c, z_c, t)$ (#) is the total number of residual particles within the space at time $t$. $V$ ($m^3$) is the volume of the compartmental space. $\dot{M}$ ($\mu g \ s^{-1}$) is the actual mass emission rate of an indoor source. $\dot{S}$ (# $s^{-1}$) is the arbitrary particle number emission rate defined by us. In our simulation, the compartmental space was defined as a sphere centered at the position of the receptor with a radius of $r$ (m) to represent the air bulk to which a person is exposed. The larger the size of the sphere, the less variability or numerical error in modeled exposure will be seen. We chose 0.05 m as the radius of the spherical compartmental space. This was to allow reasonable comparisons between measured and modeled exposures (see section 4.3.2), given that this length scale for the compartmental space is comparable to that of the CO monitor used.
4.2.2 Modeling the Greater Variation of Exposure in Close Proximity to an Active Source

Given sufficient computational efforts (number of air parcels per time step and time steps of computation), one can expect that the predictions from the stochastic random walk algorithm (eq 5(a)-(c)) will converge to the deterministic or analytical solutions and will show higher exposures near an active source. However, the algorithm does not capture the greater variability of exposure near the source that results from the random occurrences and durations of transient directional air movements of turbulent mixing indoors.

As an initial effort to capture this variability in the model without introducing additional air flow parameters and assumptions, we separate each of the equations (eq 5(a)-5(c)) into two piecewise functions to allow random sampling from either the negative or the positive half of the normal distribution, \( N(\mu = 0, \sigma^2 = 2K\Delta t) \). For example, eq 5(a) is formulated as:

\[
X^n = X^{n-1} + (2K_\alpha \Delta t)^{1/2} [\alpha] \quad \text{at} \quad p = 0.5; \ n = 1, 2, \ldots \quad (9a)
\]

\[
X^n = X^{n-1} + (2K_\alpha \Delta t)^{1/2} [-\alpha] \quad \text{at} \quad p = 0.5; \ n = 1, 2, \ldots \quad (9b)
\]

For eq 9(a) and 9(b), \(|\alpha|\) is the absolute value of \(\alpha\), representing the positive half of the unit normal distribution, \(N(\mu = 0, \sigma^2 = 1)\), while \(-|\alpha|\) represents the negative half of the unit normal distribution.

For each time step, instead of randomly sampling displacements from the whole distribution, we first incorporate a (50/50% chance) pre-selection step to determine which half of the distribution is used, applying this to all 3 directions, with mutually independent pre-selection decisions. Thus, all air parcels will move towards the same corner of the room for a given time step, but each will have a different magnitude of motion randomly chosen for each of the 3 coordinates. The resulting transient
directionality is intended to resemble what is seen in indoor environments in the presence of turbulent mixing. In the next time step, for each of the 3 coordinates, there is 50% chance that all air parcels will either maintain or reverse their original movement direction along that axis. Based on this formulation, we aim to model stochastically the random occurrences and durations of transient directional air motions within the Fickian diffusion framework for those cases where the expected values of the air parcel positions equal 0 (no time-averaged advective displacement).

4.3 MODEL VALIDATIONS

4.3.1 Comparison with Analytical Predictions

To test how well the indoor stochastic model can describe anisotropic diffusion for a thermally stratified indoor space, we defined a hypothetical 5×4×4 m room where we introduced an instantaneous release of air parcels at the center. The horizontal turbulent diffusion coefficient \( K_h = 0.01 \text{ m}^2 \text{ s}^{-1} \) was assumed to be twice as large as the vertical value \( K_v = 0.005 \text{ m}^2 \text{ s}^{-1} \). This anisotropic assumption was based in part on the observations in a house by Acevedo-Bolton et al. (2010). In this simulation, 10,000 parcels were released at \( t = 0 \) and underwent diffusion with time without any removal \( (\text{ACH} + k = 0 \text{ s}^{-1}) \). Based on the Einstein diffusion equation (eq 10), it is theoretically expected that, before the air parcels reach the 6 wall boundaries, the variance of air parcel positions should grow linearly with time, with a coefficient equal to 2 times of the turbulent diffusion coefficient \( K \).

\[
\sigma^2 = 2Kt
\]  

(10)

Using the particle tracking equations, (eq 5(a)-5(c)) along with the wall reflection algorithms, Figure 4.1 plots variances \( \sigma^2 \) of the modeled air parcel positions versus time (with time step of 1 s) for \( x \), \( y \), and \( z \)-coordinates, respectively. For each coordinate, the variance increased with time, and ultimately reached a maximum value indicating that the well-mixed state had been reached. This validates the effectiveness of the wall reflection
algorithm in preventing air parcels from escaping the space. During the initial 30-s period, all the 3 variances grow linearly with time ($R^2 > 0.999$). The slopes of regression lines for the $x$- and $y$-coordinates agreed with each other (0.02 m$^2$ s$^{-1}$), and were twice as steep as that for the $z$-coordinate (0.01 m$^2$ s$^{-1}$). This is consistent with the theoretical expectation, given that the turbulent diffusion coefficient in the $x$- and $y$-directions ($K_h = 0.01$ m$^2$ s$^{-1}$) was twice as large as that in the $z$-direction ($K_v = 0.005$ m$^2$ s$^{-1}$), and that the slope should equal $2K$. The maximum variances of the $y$- and $z$-coordinates (~1.33) converged with each other, as expected due to the same distance between 2 wall boundaries (4 m). They were smaller than that of the $x$-coordinate (~2.08), which had a longer boundary distance (5 m).

![Figure 4.1](image)

**Figure 4.1** Model simulation testing the variances of air parcel positions ($X, Y, Z$) as a function of time for an initial instantaneous release of 10,000 air parcels that were diffused anisotropically in a hypothetical 5×4×4 m room with horizontal and vertical turbulent diffusion coefficients of 0.01 and 0.005 m$^2$ s$^{-1}$, respectively.
To examine how well the our pollutant removal and wall reflection algorithms can maintain the appropriate mass balance for the system, we also modeled a continuous source at the center of the hypothetical 5×4×4 m room with the same anisotropic diffusion condition ($K_h = 0.01 \text{ m}^2 \text{s}^{-1}$ and $K_v = 0.005 \text{ m}^2 \text{s}^{-1}$). The source released an air pollutant over 1 h at a constant mass emission rate (1 $\mu\text{g} \text{s}^{-1}$), followed by another 1 h without any emission. We considered 2 extreme cases for pollutant removal: one with a zero removal rate ($ACH + k = 0 \text{ h}^{-1}$) and another with a very high removal rate ($ACH + k = 36 \text{ h}^{-1}$). Based on the first-order removal formulation (eq 6), the rate of change in the total pollutant mass ($M$) inside the room should satisfy eq 11.

$$\frac{dM}{dt} = q - (ACH + k)M$$  \hspace{1cm} (11)

Figure 4.2(a) and 4.2(b) plot the total mass of air pollutant in the room as a function of time during the initial 1-h source period and the subsequent 1-h no-emission period, for $ACH + k = 0 \text{ h}^{-1}$ and 36 $\text{h}^{-1}$, respectively. The bold grey solid lines indicate the analytical predictions from the time integration of eq 11, while the other 3 lines show the model simulation results for 3 different time steps of computations (1, 10, and 100 s). In the absence of pollutant removal (Figure 4.2(a)), $M$ increased linearly with time at rate of 1 $\mu\text{g} \text{s}^{-1}$ over the first 1-h period, maintaining a maximum value of 3,600 $\mu\text{g}$ over the second 1-h period. All 3 time steps of computations converged exactly to analytical predictions, again showing mass conservation due to the introduction of the wall boundary condition. In the presence of pollutant removal (Figure 4.2(b)), the simulation results showed a first order behavior during both the rise and decay periods, and agreed with the analytical prediction reasonably well when the time step of 1 s was used. This result indicates that by using a time step sufficiently smaller than the time scale of pollutant removal ($\tau = 100$ s), the temporal removal algorithm (eq 6) is equivalent to the analytical expectation (eq 11). Provided that the typical removal rates for natural ventilation conditions are at least an order of magnitude smaller than 36 $\text{h}^{-1}$ (Howard-Reed et al, 2002), a time step of 10 s can be used to resolve the pollutant removal with reasonable accuracy.
Figure 4.2. Model simulation tracking the total mass of the air pollutant in the hypothetical 5×4×4 m room as a function of time during an initial 1-h source period and a subsequent 1-h no-emission period for (a) zero removal rate ($ACH + k = 0 \text{ h}^{-1}$) and (b) high removal rate ($ACH + k = 36 \text{ h}^{-1}$), respectively. The bold grey solid lines indicate the analytical predictions from the time integration of eq 11, whereas the other 3 lines show the model simulation results for 3 different time steps of computations (1, 10, and 100 s).
A major purpose of this model is to predict the exposure (time-averaged concentration) as a function of the distance from an active indoor emission source. For this, we define a point source continuously releasing an air pollutant at a constant mass emission rate (1 μg s\(^{-1}\)), at the center of an infinitely large space (wall reflection can be neglected). By assuming isotropic diffusion (\(K_h = K_v = 0.0025\text{ m}^2\text{s}^{-1}\)) and no-removal (\(ACH + k = 0\text{ h}^{-1}\)) conditions, we can compare the stochastic simulation results with the corresponding deterministic predictions—the time integration of eq 1 over an exposure time, \(T\), divided \(T\).

In our simulation, we defined 4 spheres, each 0.1 m in diameter, centered at (0.25, 0, 0), (0.5, 0, 0), (1, 0, 0), and (2, 0, 0) to evaluate the exposures at 0.25, 0.5, 1, and 2 m from the source (located at the origin) over 10 min. Using a time step of 1 s and 1000 air parcels per time step, Figure 4.3(a) and 4.3(b) plots the results of 1000 repetitive runs of model simulations (box plots) compared with the deterministic or analytical predictions (dash lines), using the original (eq 5(a)-5(c)) and the new piecewise sampling algorithms, respectively. For both algorithms, the means of the 1000 simulation results converged to the deterministic predictions for the 4 different distances reasonably well, with an absolute relative difference averaged over the 4 distances (\(E\)) of less than 0.5%. Using the same numerical settings, we further tested the new algorithm for \(K\) values of 0.001, 0.01, and 0.025 m\(^2\) s\(^{-1}\), and found consistently satisfactory results (\(E < \sim 1\%) (see Table 4.1). This indicates that the new piecewise algorithm is equivalent to the original algorithm in predicting the expected exposure as a function of the distance from an active indoor source.

Compared to the original algorithm, the piecewise algorithm produces a wider range of possible exposure cases, each with an expected value that converges to the deterministic prediction. However, as the distance from the source decreases, the variations of exposures greatly increase for the piecewise algorithm. This is consistent with the observations reported from indoor tracer proximity experiments in 2 homes (Acevedo-Bolton et al., 2010).
Figure 4.3. Comparison between the mean (horizontal dashed lines in the box plots) of 1000 model simulation results of 10-min time-averaged concentration and the corresponding deterministic predictions at distances of 0.25, 0.5, 1, and 2 m from the source, using the (a) original and (b) new piecewise sampling algorithms, respectively.
Table 4.1 Comparison of the means of 1000 simulation results of 10-min exposure using the new piecewise sampling algorithm with the corresponding analytical predictions at 4 distances from the source (0.25, 0.5, 1, and 2 m), for isotropic turbulent diffusion coefficients (\(K\)) of 0.001, 0.0025, 0.01, and 0.025 m\(^2\) s\(^{-1}\).

<table>
<thead>
<tr>
<th>(K) (m(^2) s(^{-1}))</th>
<th>10-min Exposure (µg m(^{-3}))</th>
<th>(E^{(3)}) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Simulation average (^{(1)})</td>
<td>Analytical (^{(2)})</td>
</tr>
<tr>
<td></td>
<td>0.25 m</td>
<td>0.5 m</td>
</tr>
<tr>
<td>0.001</td>
<td>219.62</td>
<td>73.06</td>
</tr>
<tr>
<td></td>
<td>(218.36)</td>
<td>(72.55)</td>
</tr>
<tr>
<td>0.0025</td>
<td>100.76</td>
<td>39.29</td>
</tr>
<tr>
<td></td>
<td>(100.73)</td>
<td>(39.31)</td>
</tr>
<tr>
<td>0.01</td>
<td>28.08</td>
<td>12.57</td>
</tr>
<tr>
<td></td>
<td>(28.38)</td>
<td>(12.59)</td>
</tr>
<tr>
<td>0.025</td>
<td>11.66</td>
<td>5.55</td>
</tr>
<tr>
<td></td>
<td>(11.84)</td>
<td>(5.50)</td>
</tr>
</tbody>
</table>

\(^{(1)}\) Mean of the 10-min time-averaged concentrations from 1000 repetitive model simulations using the new piecewise sampling algorithm.

\(^{(2)}\) Analytical predictions from the time integration of eq 1 over 10 minutes, divided by 10 minutes.

\(^{(3)}\) Mean absolute relative difference (\(E\)) between the means of the 1000 model simulation results (Simulation average) and the corresponding analytical predictions (Analytical): 
\[
E = \frac{1}{4} \sum_{i=1}^{4} \left| \frac{\text{Simulation average}_i - \text{Analytical}_i}{\text{Analytical}_i} \right|
\]
Figure 4.4(a)-4.4(d) plot an example of the 10-min concentration time series at 0.25, 0.5, 1, and 2 m from the source computed by the original and the new piecewise sampling algorithms, respectively. The original sampling algorithm gives curves (dotted lines) that show only small fluctuations. The new piecewise algorithm produces much larger fluctuations in concentration (solid lines), with random occurrences and durations of spikes due to the introduced transient directionality of air parcel transport. This simulation result is similar to the occurrences of microplumes observed close to indoor active emission sources (Furtaw et al., 1996; McBride et al., 1999; McBride, 2002; Klepeis et al., 2007, Klepeis et al., 2009). As the distance from the source decreases, the magnitudes of the spikes increase significantly. These sporadic high concentration spikes contribute to the greater variation of overall (10-min) exposures in close proximity to the source (Figure 4.3(b)).
Figure 4.4. An example of the modeled 10-min concentration time series computed by the original (dotted lines) and the new piecewise sampling (solid lines) algorithms, at distances of (a) 0.25, (b) 0.5, (c) 1, and (d) 2 m from the source.
4.3.2 Comparison with Experimental Measurements

To test how well the new piecewise sampling algorithm can predict distributions of real exposure cases in the indoor environment, we selected 3 CO tracer experiments (9-2-08, 11-6-08 (night), and 11-7-08 (night)) with comparable turbulent diffusion coefficients (0.0019, 0.0023, and 0.0020 m\(^2\) s\(^{-1}\)) and source emission rates (~406 µg s\(^{-1}\)), based on the results presented in Chapter 3 (Table 3.4). This allows us to combine the time-averaged CO measurements at a given distance to the source from the 3 experiments to create a larger dataset of exposure cases measured under comparable mixing conditions. We focused on measurements at 4 different distances from the source (0.25, 0.5, 1, and 2 m). For each distance, we treated the time-averaged measurement at each angle with respect to the source as one possible case of exposure under the assumption of horizontal isotropic diffusion. Due to variations in air change rate (ACH) and wall-to-source distance across the 3 experiments, we focused on the time-averaged concentrations over the initial 10-min monitoring periods to reduce the effects of different removal and proximity to wall reflection conditions on the indoor CO measurements across different experiments.

Using the mean of the 3 turbulent diffusion coefficients (0.0021 m\(^2\) s\(^{-1}\)) and the source emission rate of 406 µg s\(^{-1}\), we repetitively computed 10-min exposures at (0.25, 0, 0), (0.5, 0, 0), (1, 0, 0), and (2, 0, 0) for 1,000 times without introducing pollutant removal or wall reflection. Figure 4.5 shows the comparison between modeled and measured frequency distributions of 10-min CO exposures on a normal probability graph, for the 4 different distances from the source. The filled and unfilled symbols represent modeled and measured exposures, respectively, with triangle, circle, square, and diamond respectively denoting 0.25, 0.5, 1, and 2 m distances from the source. Except for one 0.5 m measurement (~50 ppm), the simulation results covered the full ranges of measured exposures at each of the 4 distances from the source, and they showed a greater variation of exposure near the source.
Figure 4.5. Comparison between modeled and measured frequency distributions of 10-min exposure in a cumulative percentage plot, for 4 different distances from the source. The filled and unfilled symbols represent modeled and measured exposures, respectively, with triangle, circle, square, and diamond respectively denoting 0.25, 0.5, 1, and 2 m distances from the source.
At distances $\geq 1$ m, the modeled distributions predicted the measurements reasonably well, with comparable means, medians, and interquartile ranges (IQR) (see Table 4.2). However, noticeable discrepancies between measured and modeled distributions were seen at distances < 1 m. In addition to the relatively higher mean and median values, the measured distributions showed greater spreads of exposures with IQR $\sim$2-3 times as large as the simulation counterparts.

Table 4.2 Comparison of statistics between modeled and measured 10-min exposure distributions at 4 different distances from the source (0.25, 0.5, 1, and 2 m).

<table>
<thead>
<tr>
<th>Distance (m)</th>
<th>10-min Exposure</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Deterministic$^\text{(2)}$</td>
<td>Modeled$^\text{(3)}$</td>
</tr>
<tr>
<td></td>
<td>Mean [Median] (IQR)$^\text{(1)}$</td>
<td>n = 1000</td>
</tr>
<tr>
<td>0.25</td>
<td>41.6 [41.6] (11.1)</td>
<td>45.3 [50.0] (20.7)</td>
</tr>
<tr>
<td>0.5</td>
<td>15.8 [15.9] (8.2)</td>
<td>25.1 [27.1] (24.3)</td>
</tr>
<tr>
<td>1</td>
<td>4.4 [3.8] (4.5)</td>
<td>4.1 [3.2] (6.1)</td>
</tr>
<tr>
<td>2</td>
<td>0.56 [0.11] (0.68)</td>
<td>0.96 [0.20] (0.76)</td>
</tr>
</tbody>
</table>

$^\text{(1)}$ Interquartile range (IQR): the difference between the first quartile (25th percentile value) and the third quartile (75th percentile value).

$^\text{(2)}$ Deterministic predictions of 10-min exposures from the time integration of eq 1 over 10 minutes, divided by 10 minutes.

$^\text{(3)}$ 10-min exposures from 1,000 model simulations using the new piece-wise sampling algorithm for an isotropic diffusion coefficient of 0.0021 m$^2$ s$^{-1}$.

$^\text{(4)}$ Measured 10-min exposures from 3 CO tracer experiments under comparable mixing and tracer release conditions (see chapter 2), with n = 12 at distances of 0.25 and 0.5 m, and n = 28 at distances of 1 and 2 m.
One possibility is that the frequency of random change in the indoor drift direction is lower than that simulated by the time-independent toss-up formulation – this would imply that the directionality of real indoor air movement is expected to be more correlated in time. Another possibility is that the disagreement stems from the fact that eddy diffusion indoors is not perfectly isotropic: the vertical turbulent diffusion coefficient should be smaller than the horizontal value due to thermal stratification indoors. Having more concentrated pollutant levels within the measurement plane (1 m from the ground, equal to the source emission height) would make the magnitudes of the close-proximity concentration spikes higher than those predicted by the isotropic modeling simulation. Longer durations and/or higher concentrations of spikes may have contributed to greater spreads of observed exposures in close proximity to the source. As another consideration, this could be due in part to the inherent variations in the turbulent diffusion coefficient and source emission rate across the 3 experiments.

To summarize, the current formulation of the piecewise sampling algorithm has captured greater variability of exposure close to the source and predicted distributions of 10-min averages at distances $\geq 1$ m.
4.4 CONCLUSIONS AND IMPLICATIONS

To model close proximity exposure to an indoor active air pollution point source, an indoor exposure model has been developed, invoking the random-walk particle tracking method. The proposed model considers: (i) anisotropic indoor eddy diffusion; (ii) a continuous point source release; (iii) the first-order removal of air pollutant due to indoor-outdoor air exchange and surface deposition/adsorption; and (iv) the air pollutant reflection from wall boundaries. The important feature of the model is that it is suited to indoor spaces without pronounced and sustained advection (i.e. in the absence of operating HVAC system and fans), which applies to most of the residential indoor environments. Using this model, exposures at indoor locations of interest can be estimated based on the following variables: source emission rate; horizontal and vertical turbulent diffusion coefficients; air change and surface removal rates; positions of the point source, receptors, and walls; and emission and exposure durations.

To model the highly variable exposure near an active source, we developed a new piecewise random walk algorithm to stochastically simulate the transient directional movements of air pollutants due to turbulent mixing indoors. We found that this new algorithm can produce a wide range of different exposure cases while preserving ensemble averages that satisfy the principle of Fickian diffusion. The simulation results reasonably covered the wide range of measured values and captured greater variation of exposure in close proximity to the source; however, it underestimated the range of variation at proximities < 1 m from the source.

Further model modifications could be made to more realistically delineate the transient directional air motions of turbulent mixing indoors (i.e. autocorrelated direction of air motions). The presented modeling concept could be a new starting point in predicting transient peak exposures (exposures to microplumes) close to an active source under turbulent mixing conditions, not only for indoors but also for other environmental applications.
REFERENCES


5.1 MAJOR FINDINGS/CONTRIBUTIONS

This thesis study was the first effort to characterize and model theoretically the proximity effect for exposures in residential environments. Along the way, I developed a signal reconstruction method to accurately measure CO concentrations near a source. The major findings/contributions from this work are as follows:

1) I developed a theoretical model based on Fick’s Law to reconstruct accurate concentration time series from monitor readings for CO and other diffusion-limited gas sensors. Results showed that this model can reconstitute transient profiles of rapidly-varying input concentrations with reasonable accuracy. This method is useful for applications aiming to measure short-term peak exposures (i.e. acute exposures to toxic gases) or examine transient turbulent mixing characteristics in close proximity to a source.

2) Using our indoor monitor array (30-37 sensors), I found that the isotropic turbulent diffusion formulation used in occupational exposure models can reasonably describe exposures as a function of horizontal distance from the source in natural ventilated residences, over an averaging time scale of 30 min. This was the first attempt to test the validity of the existing proximity exposure models using a massive monitor array, and the first involving a residential setting.

3) I determined the first set of experimental estimates of turbulent diffusion coefficients in naturally ventilated residential rooms, finding that they ranged from 0.001 to 0.015 m² s⁻¹ for air change rates of 0.2-5.4 h⁻¹. These values will allow the use of currently available isotropic eddy diffusion models to predict the effect of
proximity on personal exposure in the presence of an in-home air pollution point source.

4) Representing the air mixing rate as the turbulent diffusion coefficient divided by the square of the indoor volume length scale, I was able to normalize for variations in room size and found a significant overall positive linear correlation between the air change rates and the air mixing rates for the two rooms combined. This suggests a possible new approach for predicting the turbulent coefficient for an indoor space of interest, using just 2 readily-measured parameters: air change rate and the dimensions of the indoor space.

5) To model the high variability in exposure near an active source, I developed a new piecewise random walk algorithm to stochastically simulate the transient directional air movements of turbulent mixing indoors. This new algorithm produced a wide range of different exposure cases, while preserving ensemble averages that satisfy the principle of Fickian diffusion. The presented modeling concept offers a new starting point for predicting transient peak exposures close to an active source under turbulent mixing conditions, with potential applications not only involving indoors but also other environmental locations.

5.2 FUTURE RESEARCH

The findings presented in this dissertation from both the characterization and modeling studies suggest additional research avenues. Based on my findings, I suggest the following topics for possible future investigation:

1) Our indoor monitor array study was focused on measuring the horizontal distribution of exposure at breathing height and examined how well the horizontal concentration profiles can be described by a single isotropic turbulent diffusion coefficient, as used by currently available analytical proximity exposure models. It would be worthwhile to further examine both horizontal and vertical
concentration profiles to test under what circumstances the isotropic formulation can be a reasonable approximation and under what circumstances an anisotropic model should be used instead, based on the magnitude of indoor thermal stratification.

2) In the monitor array experimental study, I found a consistent linear relationship between air mixing rate and air change rate from the 2 rooms. It would be valuable to conduct follow-up experiments to examine how generalizable this relationship is for other naturally-ventilated indoor spaces of interest.

3) In the monitor array experimental study, we examined how the indoor-outdoor air exchange (a kinetic energy source) affects the magnitude of turbulent mixing indoors. It would be worthwhile to conduct parallel studies investigating how thermal energy sources, such as incoming solar radiation and an in-home space heater, contribute to turbulent diffusion indoors, and what the combined effect of the 2 different types of energy inputs would be on indoor air pollutant dispersion.

4) To account for the high variability in exposure close to the source, I developed a new piecewise sampling algorithm to simulate the transient directionality of turbulent mixing indoors. However, the current time-independent formulation of the model doesn’t fully capture the variability of exposures within 1 m from the source. It would be valuable to further test this piecewise modeling approach with the incorporation of auto-correlated directional air flow (i.e. random walk correlated in time) to more realistically capture the magnitudes and time scales of concentration fluctuations close to the source.
APPENDIX A

MATLAB script of the random-walk indoor exposure model

%%% < INPUT VARIABLES > %%%

%%Source%%
Fem=1; % source mass emission rate(ug/s)
T=600; % emission duration(s)
Xo=0; % source position x-coordinate (m)
Yo=0; % source position y-coordinate (m)
Zo=0; % source position z-coordinate (m)

%% Turbulent diffusion coefficient %%
K_h=0.001; % horizontal (m^2/s)
K_v=0.001; % vertical (m^2/s)

%% Removal rate %%
Rv=0/3600; % Rv = ACH+k (1/s)

%% Wall positions %%
Xwall_n=-1000; % (m)
Xwall_p=1000; % (m)
Ywall_n=-1000; % (m)
Ywall_p=1000; % (m)
Zwall_n=-1000; % (m)
Zwall_p=1000; % (m)

%% Receptors %%
xr=[0.25,0.5,1,2]; % Receptor position x-coordinate (m)
yr=[0,0,0,0]; % Receptor position y-coordinate (m)
zr=[0,0,0,0]; % Receptor position z-coordinate (m)
T_exposure=600; % Exposure duration(s)

choice='original'; % choose 'original' or 'new' algorithm

%% Numerical property %%
N=1000; % number of air parcels released per time step (#)
deltat=1; % time step(s)
r=0.05; % radius of the exposure sphere (m)

%~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~%
X=Xo*ones(N,1); Y=Yo*ones(size(X)); Z=Zo*ones(size(X));

R=100*ones(size(X)); % parallel array for removal (m)

X1=X; Y1=Y; Z1=Z; R1=R;
S=zeros(size(xr));

n=0;

while n*deltat<T_exposure;
    n=n+1;
    switch(choice)
    case 'original'
        eta1=randn(size(X)); eta2=randn(size(Y)); eta3=randn(size(Z));
    case 'new'
        a=rand-0.5; b=rand-0.5; c=rand-0.5;
        if a>=0;
            eta1=abs(randn(size(X)));
        else a<0;
            eta1=-abs(randn(size(X)));
        end
        if b>=0;
            eta2=abs(randn(size(Y)));
        else b<0;
            eta2=-abs(randn(size(Y)));
        end
        if c>=0;
            eta3=abs(randn(size(Z)));
        else c<0;
            eta3=-abs(randn(size(Z)));
        end
    end
    X=X+eta1*(2*K_h*deltat)^0.5;
    Y=Y+eta2*(2*K_h*deltat)^0.5;
    Z=Z+eta3*(2*K_v*deltat)^0.5;
    X(find(X>=Xwall_p))=2*Xwall_p-X(find(X>=Xwall_p));
    X(find(X<=Xwall_n))=2*Xwall_n-X(find(X<=Xwall_n));
    Y(find(Y>=Ywall_p))=2*Ywall_p-Y(find(Y>=Ywall_p));
    Y(find(Y<=Ywall_n))=2*Ywall_n-Y(find(Y<=Ywall_n));
    Z(find(Z>=Zwall_p))=2*Zwall_p-Z(find(Z>=Zwall_p));
    Z(find(Z<=Zwall_n))=2*Zwall_n-Z(find(Z<=Zwall_n));
    if n*deltat<T;
        X(:,1:n)=X; X(:,n+1)=X1;
        Y(:,1:n)=Y; Y(:,n+1)=Y1;
        Z(:,1:n)=Z; Z(:,n+1)=Z1;
    else
        X=X; Y=Y; Z=Z;
    end
end
R(:,1:n)=R*exp(-Rv*dt); R(:,n+1)=R1;
else
R=R*exp(-Rv*dt);
end
%%% concentration interpretation %%%
for i=1:length(xr);
    N(i)=sum(R(find(sqrt((X-xr(i)).^2+(Y-yr(i)).^2+(Z-zr(i)).^2)<=r)));
    C(i)=(((Fem*dt)/sum(R1))*N(i))/(pi*(4/3)*r^3); %ug/m^3
end
S=S+C;
end
Exposure=S/n %ug/m^3
APPENDIX B

Association of Size-Resolved Airborne Particles with Foot Traffic Inside a Carpeted Hallway

Kai-Chung Cheng, Marian D. Goebes, Lynn M. Hildemann

ABSTRACT
The effect of foot traffic on indoor particle resuspension was evaluated by associating non-prescribed foot traffic with simultaneous size-resolved airborne particulate matter (PM) concentrations in a northern California hospital. Foot traffic and PM were measured every 15 min in a carpeted hallway over two 27-hr periods. The PM concentration in the hallway was modeled based on the foot traffic intensity, including the previous PM concentration via an autocorrelation regression method based on the well-mixed box model. All 5 size ranges of PM, ranging from 0.75-1µm to 5-7.5µm, were highly correlated with foot traffic measurements for both monitoring periods ($p < 0.001$, $R^2 = 0.87-0.90$). However, correlations during daytime hours were less significant than nighttime. Coefficients found via this autoregressive analysis can be interpreted to reveal (i) time-independent contributions of walking activities on PM levels for a specific location; and (ii) size-specific characteristics of the resuspended PM.

6 Published in *Atmospheric Environment*, 2010, 44, 2062-2066. Reproduced by permission of Elsevier.
B.1. INTRODUCTION

Particle resuspension due to walking activities has been identified as an important contributor to airborne particulate matter (PM) in indoor environments. Thatcher and Layton (1995) found that walking in a carpeted room increased PM levels by 100% for some supermicron particles in a California residence. Long et al. (2000) surveyed 9 Boston-area homes, finding that vigorous walking contributed 12µg/m$^3$ to indoor PM$_{2.5}$ concentrations. Ferro et al. (2004a) found that one person walking on carpet contributed 15µg/m$^3$ to PM$_{2.5}$ concentrations in a California house. Qian et al. (2008) reported that a walking activity period elevated concentrations of PM$_{10}$ by 37µg/m$^3$ in a residence.

Many laboratory studies have examined factors that potentially influence the contributions of foot traffic to indoor PM levels, such as flooring types (Buttner et al., 2002), humidity and dust type (Gomes et al., 2007), dust loading (Gomes et al., 2007; Rosati et al., 2008), and the combined effect of carpet age and relative humidity (Rosati et al., 2008). However, only a few studies have investigated the effect of foot traffic intensity (number of people per unit time) on size-resolved PM levels in the field. Ferro et al. (2004a) found that a prescribed walking activity with two people produced PM$_{2.5}$ and PM$_{10}$ ~3 and 1.5× as high (respectively) as for one person. Qian et al. (2008) showed that concentrations of airborne tracer particles for a prescribed two-person walking activity were ~3× as high as one person walking. With only two levels of foot traffic considered, these results cannot be quantitatively extended to higher foot traffic levels. In addition, prescribed walking does not reflect sporadic foot traffic typical of real indoor environments.

One investigation (Luoma and Batterman, 2001) involved non-prescribed walking activities and size-resolved PM in an office space, statistically characterizing the associations of PM levels with 10 variables representing various types, locations, and lag-times of foot traffic. This study’s time-resolved measurements over many hours provided valuable variations in both foot traffic and PM. However, the complexity of the location and the variety of activities occurring (including smoking in a nearby room, standing near
the monitor, and walking by at various distances) made interpretations challenging. None of the other existing literature in this area has involved an intensive non-prescribed monitoring study.

Our study considered non-prescribed walking on a carpeted hallway in a public building. The type and proximity of foot traffic were simpler, and there were no other major PM sources. Our first goal was to statistically test how well simple models can predict size-resolved PM levels using a single variable for foot traffic. Our second goal was to use the model regressions to assess (i) the contributions of foot traffic to indoor PM levels; and (ii) size-selective characteristics of particle resuspension from walking activities.

B.2. MATERIALS AND METHODS

Research location and sampling periods. This study was carried out in a hallway of a children’s hospital in northern California. Served by an air supply system with four filters in series, the area, with relatively new commercial loop carpet, is vacuumed daily and shampooed every 1-2 weeks, with blowers used immediately afterwards to finish drying the carpet (see Goebes et al., 2008 for additional details). PM and foot traffic measurements were taken continuously, over two 27-hr weekday experiments (Mar 2007). Each was initiated within a few days after the carpet was shampooed, to provide starting conditions with conservatively low dust loadings. The relative humidity averaged 38% and 36% during the two study periods. Each period included regular clinic (8am–5pm), late clinic (6pm–9pm), and off-peak (10pm–7am) hours, providing foot traffic variability.

Foot traffic and PM monitoring. Time-resolved foot traffic intensities (#/min) were measured by counting the people passing by during each 15-min interval. A Grimm Portable dust monitor Series 1.100 (Grimm Technologies, Inc., Douglasville, GA, USA) at 0.3m height measured PM number concentrations (#/m³) for 0.75-1µm, 1-2µm, 2-3.5µm, 3.5-5µm, and 5-7.5µm size ranges at 1-min resolution. Collocated gravimetric filters (Durapore membrane, diameter, 47mm; pore diameter, 0.45µm; Millipore Co.,
Billerica, MA, USA), at 0.7m height, collected PM$_6$ measurements downstream of a cyclone separator every 4 hrs. (PM$_6$ was collected to also study the behavior of *Aspergillus* mold particles, which have a maximum diameter of 6µm.) The sampling flow rate of the cyclone, measured using a Bubble Generator (Gilian Instrument, Co., West Caldwell, NJ, USA) removed particles >6.2µm. A Tinytag Ultra data logger (Gemini Data Loggers, Ltd., Chichester, UK) logged temperature and humidity every 30 seconds. For further sampling details, see Goebes *et al.* (2008).

**Data Analysis.** To estimate the 1-min PM mass concentrations, $PM_i$ (µg/m$^3$), from the number concentrations for size range $i$, particles were assumed to be spherical:

$$PM_i = N_i \left( \frac{\rho}{10^3} \right) \left( \frac{\pi D_{mi}^3}{6} \right)$$

(1)

Here, $N_i$ (#/m$^3$) is the number concentration; $\rho$ (g/cm$^3$) is the particle density, assumed to be 1; and $D_{mi}$ (µm) is the arithmetic midpoint of the upper and lower diameters for each size class. Then, based on the gravimetric mass measurements, the eq.1 values were multiplied by a factor of 2.04 – this was the slope from a linear regression between the 4-hr gravimetric PM$_6$ measurements and the corresponding time-integrated mass estimates based on the particle counts ($R^2$=0.95, n=14). (Linearly-interpolated 5-6µm concentrations within the 5-7.5µm size class were added to PM$_5$ to calculate PM$_6$.)

Further details of this methodology are discussed in Ferro (2002), and Ferro *et al.* (2004a). This rescaling factor may reflect an actual particle density greater than 1, and/or average diameters somewhat greater than our assumed midpoint values.

*The direct regression model:* To explore the relationship between foot traffic intensities and size-resolved PM levels, 15-min time-averaged concentrations were calculated for each size range, and correlated linearly with the simultaneous foot traffic intensities as:

$$C_i^n = \beta_0 + \beta_1 (FT^n)$$

(2)
In this model, $C_i^n$ is the $n$th time-averaged concentration for size class $i$ ($\mu g/m^3$); $FT^n$ is the concurrent foot traffic intensity (#/min); and $\beta_o$ and $\beta_1$ are correlation coefficients.

The “autocorrelative regression model”: For time series measurements where indoor sources vary with time, the influence of previous airborne concentrations on subsequent measurements can be substantial (Luoma and Batterman, 2000). To capture this autocorrelation effect, it can be assumed that concentrations decay in an exponential fashion (based on the well-mixed box model) and contribute to the background of subsequent measurements (Ferro et al., 2004b). Then, $C_i^n$ can be modeled by adding the current foot traffic contribution with the exponentially-decaying previous airborne concentration:

$$C_i^n = \beta_o + \beta_1(FT^n) + C_i^{n-1} \exp(-k_i \Delta t)$$

(3)

In eq 3, $C_i^{n-1}$ is the $(n-1)$th time-averaged concentration ($\mu g/m^3$) used to capture all residual source contributions; $k_i$ is a constant representing the size-specific PM removal rate (1/min); and $\Delta t$ is the time interval between two consecutive measurements (15 min for our study). Since $k_i$ and $\Delta t$ can be treated as constants for each size class of particles, this autoregressive regression model can be simplified to:

$$C_i^n = \beta_o + \beta_1(FT^n) + \beta_2 C_i^{n-1}$$

(4)

This form of the autocorrelation coefficient, $\beta_2$, within an indoor air quality time series has been previously proposed for industrial hygiene applications (Roach, 1977). A similar autoregressive method was employed in Luoma and Batterman’s study (2001). But here, there is only one term for foot traffic, whereas the 2001 study used 10 terms to examine differences in the activity’s location, type (walking vs standing), and lag time. Lagged foot traffic terms were not included in eq 4 because of the longer averaging time interval (15 min) used here, and our assumption that the autocorrelation term already accounts for all preceding source contributions.
In this study, regressions using eq 2 and eq 4 were performed using the R statistical package, version 2.6.2 (http://www.r-project.org).

**B.3. RESULTS AND DISCUSSIONS**

Simultaneous PM and foot traffic measurements are plotted in Figure B.1 for the two study periods (a Monday morning through Tuesday afternoon, \( n = 105 \); and a Thursday morning through Friday afternoon, \( n = 103 \)). Both foot traffic and PM levels increased in the morning and decreased in the afternoon, and also varied in synch over shorter time scales. The one exception, a pronounced jump in PM levels observed at ~8am during the first study period (Figure B.1a), had a much larger contribution from fine PM than any other period, and coincided with a transient, concurrent increase in indoor temperature (~5.6 °F). The 3 largest PM measurements were statistically excluded as outliers \( (p<0.001, \text{Bonferroni Outlier Test}) \).
Figure B.1 Size-specific PM levels and their simultaneous foot traffic measurements. Each observation represents a 15-min averaged measurement. (a) First study period from Monday-10:30 to Tuesday-14:00 in March 2007; (b) Second study period from Thursday-10:30 to Friday-14:00 in March 2007.
Table B.1 summarizes statistics for PM\textsubscript{2.5}, PM\textsubscript{7.5}, and foot traffic intensity for the two study periods. The distributions of measurements were neither normal nor log-normal ($p<0.01$, Shapiro-Wilks test), so means, medians, 25\textsuperscript{th} percentile (Q1) and 75\textsuperscript{th} percentile (Q3) values are given. The large interquartile ranges (where IQR=Q3-Q1) obtained for both foot traffic and PM reflect the order of magnitude variation seen in these measures. Elevations in dust reservoir levels in the carpet (e.g., due to measuring right after a weekend) could have contributed to the substantially higher Q3 values for PM seen in the first study period.

Table B.1 Statistics of 15-min averaged measurements for foot traffic, PM\textsubscript{2.5}, and PM\textsubscript{7.5} in the two study periods.

<table>
<thead>
<tr>
<th>Study Period</th>
<th>$n$\textsuperscript{(1)}</th>
<th>$FT$ (#/min)\textsuperscript{(2)}</th>
<th>PM Concentration ($\mu g/m^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Mean [Median] (Q1,Q3\textsuperscript{(5)})</td>
<td>Mean [Median] (Q1,Q3\textsuperscript{(5)})</td>
</tr>
<tr>
<td>First</td>
<td>102\textsuperscript{(4)}</td>
<td>3.16 [2.73] (0.47,5.27)</td>
<td>3.50 [1.65] (0.44,5.79) (4.22,40.69)</td>
</tr>
<tr>
<td>Second</td>
<td>103</td>
<td>3.08 [2.93] (0.42,5.44)</td>
<td>2.50 [1.84] (0.60,3.61) (5.15,32.08)</td>
</tr>
</tbody>
</table>

\textsuperscript{(1)} $n$ = number of observations.
\textsuperscript{(2)} $FT$ = foot traffic intensity measured by counting number of people passing the monitoring location every 15 min.
\textsuperscript{(3)} PM\textsubscript{2.5} was estimated by adding linearly interpolated 2-2.5 $\mu m$ concentration within 2-3.5 $\mu m$ size class to PM\textsubscript{2}.
\textsuperscript{(4)} Excludes 3 outlier measurements observed at ~8am.
\textsuperscript{(5)} Q1 = first quartile (25th percentile value); Q3 = third quartile (75th percentile value).

In Table B.2, combined data from the 2 study periods are sorted into daytime (7am–7pm) vs. nighttime (7pm–7am) – this grouped the data into higher vs. lower foot traffic periods. For all size fractions, the mean PM concentrations during the higher foot traffic periods were $\sim$5-7× as high as during the lower foot traffic periods. The coarser PM ($\geq 2\mu m$) was 93-95% of the total average PM\textsubscript{7.5} mass during both day and night.
Using the direct regression model (eq.2) for all data combined, the correlation between foot traffic intensities and PM levels was significant (Table B.3) for all particle size ranges ($p<0.001$). The $R^2$ values systematically increased, from 0.53 to 0.70, as particle size increased. This is not surprising – since larger particles more quickly redeposit after being suspended, a model based solely on emission strength should become more strongly predictive as particle size increases.

For the autoregressive model (eq.4), both foot traffic intensities and previous concentration measurements showed strong correlations (Table B.3) with all size ranges of PM ($p<0.001$). Substantially higher $R^2$ values were obtained, ranging from 0.87 to 0.90. The $R^2$ values obtained here for PM$\geq 1\mu$m are much higher than the values reported by Luoma and Batterman (2001) for their autoregression analyses – this is likely due in large part to the greater simplicity of our field site.
For the autoregressive model, the smallest size range of PM showed the largest $R^2$ values, and the largest correlation coefficients for previous concentrations – these trends are likely due to the longer persistence of small airborne particles. The coefficients for foot traffic increased as particle size increased, indicating that the bulk of PM resuspended from the carpet was coarse. This result agrees with previous findings that particle resuspension due to indoor walking activities was most pronounced for supermicron particles (Thatcher and Layton, 1995; Abt et al., 2000; Long et al., 2000; Ferro et al., 2004). However, it does not imply that larger-sized particles are more easily resuspended, as this study did not collect information on the relative amounts of different-sized particles deposited in the carpeting.

Measurements were further divided into daytime (high foot traffic) versus nighttime (low foot traffic). The coefficients obtained using the autocorrelative regression model (see Table B.3) exhibited size-specific magnitudes and trends for both groups that were, in general, comparable to that for all data combined. Correlations between previous concentration components and PM levels remained highly significant ($p < 0.001$).

However, the correlations between foot traffic and PM levels for the high foot traffic group were not as significant as for the low foot traffic group. We hypothesize that early high foot traffic events might deplete most of the resuspendable particles on the carpet, resulting in less PM resuspended during later periods of comparably high foot traffic. Some support for this can be seen in the 5-7.5$\mu$m PM, where both groups showed significant correlations with foot traffic intensities ($p < 0.01$). The foot traffic coefficient for the low foot traffic group was $>2\times$ as high as for the high foot traffic group, consistent with the expectation that the carpet’s dust loading should be higher at nighttime than in the daytime. A doubling in the foot traffic coefficient was also seen at night for the 3.5-5$\mu$m PM, although the significance of the daytime correlation with foot traffic was weaker ($p < 0.05$).
Table B.3 Correlation statistics of the direct regression model and the autocorrelative regression model.

<table>
<thead>
<tr>
<th>Particle Size</th>
<th>0.75-1µm</th>
<th>1-2 µm</th>
<th>2-3.5 µm</th>
<th>3.5-5 µm</th>
<th>5-7.5 µm</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Direct Regression Model</strong>&lt;sup&gt;(2)&lt;/sup&gt;</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>All Foot Traffic Intensity (n = 201)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\beta_1$</td>
<td>0.104 ***</td>
<td>0.417 ***</td>
<td>1.27 ***</td>
<td>2.13 ***</td>
<td>2.75 ***</td>
</tr>
<tr>
<td>$\beta_o$</td>
<td>0.007</td>
<td>0.00150</td>
<td>0.102</td>
<td>0.715</td>
<td>1.69**</td>
</tr>
<tr>
<td>$R^2$</td>
<td>0.534</td>
<td>0.554</td>
<td>0.583</td>
<td>0.648</td>
<td>0.697</td>
</tr>
<tr>
<td><strong>Autocorrelative Regression Model</strong>&lt;sup&gt;(3)&lt;/sup&gt;</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>All Foot Traffic Intensity (n = 201)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\beta_1$</td>
<td>0.0254 ***</td>
<td>0.104 ***</td>
<td>0.330 ***</td>
<td>0.681 ***</td>
<td>1.04 ***</td>
</tr>
<tr>
<td>$\beta_2$</td>
<td>0.709 ***</td>
<td>0.706 ***</td>
<td>0.698 ***</td>
<td>0.649 ***</td>
<td>0.598 ***</td>
</tr>
<tr>
<td>$\beta_o$</td>
<td>0.00459</td>
<td>0.0123</td>
<td>0.0673</td>
<td>0.275</td>
<td>0.672</td>
</tr>
<tr>
<td>$R^2$</td>
<td>0.901</td>
<td>0.893</td>
<td>0.883</td>
<td>0.878</td>
<td>0.867</td>
</tr>
<tr>
<td>Nighttime Foot Traffic Intensity less than 3 (n = 104)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\beta_1$</td>
<td>0.00825 *</td>
<td>0.0548 **</td>
<td>0.318 **</td>
<td>1.01 ***</td>
<td>1.89 ***</td>
</tr>
<tr>
<td>$\beta_2$</td>
<td>0.816 ***</td>
<td>0.772 ***</td>
<td>0.680 ***</td>
<td>0.544 ***</td>
<td>0.474 ***</td>
</tr>
<tr>
<td>$\beta_o$</td>
<td>0.00833</td>
<td>0.0258</td>
<td>0.0762</td>
<td>0.192</td>
<td>0.340</td>
</tr>
<tr>
<td>$R^2$</td>
<td>0.818</td>
<td>0.794</td>
<td>0.746</td>
<td>0.732</td>
<td>0.716</td>
</tr>
<tr>
<td>Daytime Foot Traffic Intensity greater than or equal to 3 (n = 97)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\beta_1$</td>
<td>0.0248 *</td>
<td>0.0955</td>
<td>0.293</td>
<td>0.570 *</td>
<td>0.802 **</td>
</tr>
<tr>
<td>$\beta_2$</td>
<td>0.704 ***</td>
<td>0.701 ***</td>
<td>0.696 ***</td>
<td>0.648 ***</td>
<td>0.590 ***</td>
</tr>
<tr>
<td>$\beta_o$</td>
<td>0.0138</td>
<td>0.0803</td>
<td>0.304</td>
<td>0.920</td>
<td>2.16</td>
</tr>
<tr>
<td>$R^2$</td>
<td>0.808</td>
<td>0.783</td>
<td>0.748</td>
<td>0.699</td>
<td>0.627</td>
</tr>
</tbody>
</table>

<sup>(1)</sup> $p < 0.001$ denoted by ***; $p < 0.01$ denoted by **; $p < 0.05$ denoted by *

<sup>(2)</sup> $c_i^n = \beta_o + \beta_1(FT_i^n)$

<sup>(3)</sup> $c_i^n = \beta_o + \beta_1(FT_i^n) + \beta_2 c_i^{n-1}$
Based on the daytime and nighttime foot traffic coefficients, an incremental foot traffic intensity of one person per minute in this hallway can generate sustained indoor increases of 1.0µg/m³ and 0.8µg/m³ for 1-5μm and 5-7.5μm particles, respectively, during daytime hours; increases were higher (1.4 and 1.9µg/m³, respectively) during nighttime hours.

The previous office study (Luoma and Batterman, 2001) estimated daytime increases due to walking of 0.2µg/m³ and 0.4µg/m³ for 1-5μm and 5-10μm particles, respectively (at 0.4m height); estimated increases were larger (0.7 and 1.2µg/m³) for someone spending 1 min in close proximity to the monitor.

Differences between the two studies could be due to variations in the type or age of flooring, dust loading, dust type, relative humidity, the distance between foot traffic and the monitors, the effective indoor mixing volume, the sampling height, and/or the methods used to rescale particle number counts to agree with gravimetric measurements. Thus, these quantitative estimates of PM increases are not generalizable for other indoor environments.
B.4. SUMMARY AND IMPLICATIONS

While it is well established that foot traffic can resuspend particles from carpeting, this hallway study demonstrates how major this source of indoor PM can be -- 87-90% of the variability in PM concentration was due to variations in foot traffic. Coarse PM contributed the bulk of resuspended particle mass, but persisted in the air for less time than the fine particles.

In retrospect, the following study design factors contributed to the strong statistical correlations found: (1) a hallway layout, which allowed PM to be measured in close, reproducible proximity to the sources of resuspension; (2) no significant confounding indoor sources (like cooking or smoking); (3) a ventilation system that filtered the air entering from outdoors; (4) an averaging time greater than the timescale for transport between resuspension sources and the sampler; and (5) the inclusion of an autocorrelation term to account for previous source contributions to subsequent PM measurements.

The approach demonstrated in this study could be used to determine the source strength of particle resuspension due to foot traffic when the effective indoor mixing volume can be estimated. In addition, this method is of potential value for characterizing other types of sporadic indoor source emissions. For this carpeted hospital hallway, the study demonstrated the sizable impact that foot traffic can have on indoor PM concentrations.

This methodology would allow future studies to determine which reduction strategies would be most effective. Comparisons could be made of the PM resuspended before versus after carpet shampooing. A hard plastic walking strip on top of the carpeted floor could distinguish particle emissions from occupant clothing. Such studies are particularly important for hospitals, where exposure of sensitive patients to certain types of bioaerosols can cause serious health consequences.
ACKNOWLEDGMENTS

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