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A method to measure the ozone penetration factor in residences under infiltration conditions: application in a multifamily apartment unit

Abstract Recent experiments have demonstrated that outdoor ozone reacts with materials inside residential building enclosures, potentially reducing indoor exposures to ozone or altering ozone reaction byproducts. However, test methods to measure ozone penetration factors in residences (P) remain limited. We developed a method to measure ozone penetration factors in residences under infiltration conditions and applied it in an unoccupied apartment unit. Twenty-four repeated measurements were made, and results were explored to (i) evaluate the accuracy and repeatability of the new procedure using multiple solution methods, (ii) compare results from 'interference-free' and conventional UV absorbance ozone monitors, and (iii) compare results against those from a previously published test method requiring artificial depressurization. The mean ($\pm$s.d.) estimate of P was 0.54 $\pm$ 0.10 across a wide range of conditions using the new method with an interference-free monitor; the conventional monitor was unable to yield meaningful results due to relatively high limits of detection. Estimates of P were not clearly influenced by any indoor or outdoor environmental conditions or changes in indoor decay rate constants. This work represents the first known measurements of ozone penetration factors in a residential building operating under natural infiltration conditions and provides a new method for widespread application in buildings.

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Key words: Outdoor ozone; Human exposure; Infiltration factor; Indoor chemistry; Building envelope; Building enclosure.

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Practical Implications

Ambient ozone can react with building materials as outdoor air infiltrates through cracks and gaps in exterior building enclosure assemblies. Here, we report on a method to measure ozone penetration factors in residential buildings under natural infiltration conditions and apply the test method in an unoccupied test apartment unit. Results suggest that approximately half of outdoor ozone reacts away within the building envelope in this particular apartment unit, on average, while approximately half infiltrates indoors where it can then lead to indoor exposures to ozone and ozone reaction byproducts. Results also suggest that ozone penetration factors may be lower in multifamily residences than what is typically assumed, which may alter the magnitude and type of ozone reaction byproducts found indoors.

Introduction

Elevated outdoor ozone concentrations are consistently linked with a variety of adverse health effects (Bell et al., 2004, 2006, 2014; Fann et al., 2012; Ito et al., 2005; Jerrett et al., 2009; Smith et al., 2009). Because people spend the majority of their time inside buildings (Klepeis et al., 2001) and outdoor ozone infiltrates indoors (Avol et al., 1998; Cattaneo et al., 2011; Lee et al., 2004; Liu et al., 1995; Stephens et al., 2012), cumulative indoor exposures to outdoor ozone are often larger than outdoor exposures, particularly in homes (Chen et al., 2011; Weschler, 2006). Ozone is also a primary driver of homogenous and heterogeneous chemistry in indoor environments (Fadeyi et al., 2013; Shu and Morrison, 2011; Wang and Waring, 2014; Waring, 2014; Waring and Siegel, 2013; Waring and Wells, 2014; Weschler, 2000). Because indoor exposures to both ozone and byproducts from ozone reactions with surfaces and other compounds found indoors are also of concern for health (Weschler, 2006), it is crucial to understand the mechanisms that

contribute to ozone losses in residential indoor environments.

For a given indoor environment without indoor ozone sources, the time-averaged indoor–outdoor (I/O) ozone concentration ratio (C_{in}/C_{out}, or the 'infiltration factor') is a function of the ozone penetration factor through the building enclosure and/or ventilation system (P), the air exchange rate (AER, or λ, per hour), and the first-order ozone decay rate constant (k, per hour), as shown in Equation (1).

$$\frac{C_{in}}{C_{out}} = \frac{P\lambda}{\lambda + k} \quad (1)$$

Numerous studies have measured the indoor proportion of outdoor ozone in residences under a wide variety of operational conditions (Avol et al., 1998; Brauner et al., 2014; Cattaneo et al., 2011; Lee et al., 1999; Liu et al., 1995; Romieu et al., 1998; Zhang and Lioy, 1994). In buildings that rely on natural infiltration for ventilation air, which represents the majority of residential buildings in the U.S. (Chan et al., 2005), occupants are exposed to ozone of outdoor origin (and any reaction byproducts generated indoors) only after ambient ozone penetrates indoors through cracks, gaps, and other penetrations in the building enclosure. Most previous work on indoor ozone of outdoor origin has assumed that ozone penetrates through building enclosures 100% efficiently (Chen et al., 2011; Gall et al., 2011; Georgopoulos et al., 2005; Waring, 2014; Weschler, 2000, 2006). However, limited experimental and modeling investigations have demonstrated otherwise (Liu and Nazaroff, 2001; Stephens et al., 2012; Walker and Sherman, 2013).

We previously developed a method for measuring ozone penetration factors through building enclosures and applied it in eight single-family homes, demonstrating that the mean ozone penetration factor was ~0.79, ranging from 0.62 ± 0.09 to 1.02 ± 0.15 in the small sample of residences (Stephens et al., 2012). These data combined with knowledge of reaction probabilities of a number of materials typically used in building enclosures (Liu and Nazaroff, 2001; Walker and Sherman, 2013) suggest that ozone penetration factors are likely less than unity in many residences. Lower ozone penetration factors would have important implications for both public health and building design. If ozone losses occur primarily within the enclosure, indoor exposures to ozone and ozone reaction byproducts may be lower than previously assumed and projected in a substantial number of buildings (U.S. EPA, 2007, 2014a). Thus, more field measurements of ozone penetration factors in a greater number and variety of buildings are warranted to improve our understanding of the public health risks due to indoor exposures to outdoor ozone and to enable the design of building enclosures that can intentionally scavenge outdoor ozone (Gall et al., 2011; Kunkel et al., 2010).

One key limitation in our previous experimental method is that it required the use of a large fan (i.e., blower door) to depressurize the test home to artificially elevate the AER and indoor ozone concentration above the detection limit of the UV photometric ozone monitor that was used at the time. The monitor relied on UV absorption at 254 nm, a technology that is known to have interference issues with Hg, water vapor, and a variety of indoor VOCs including styrene, methylstyrene, *o*-cresol, nitrocresol, and other aromatic species with substituted electron withdrawing groups (e.g., -OH, -NO_2, and -CHO) (ASTM D5156, 2008; Grosjean and Harrison, 1985; Huntzicker and Johnson, 1979; Johnson et al., 2014; Ollison et al., 2013; Spicer et al., 2010; Williams et al., 2006; Wilson and Birks, 2006), which may have influenced detection limits indoors and necessitated the use of the blower door method. Further, the reliance on artificially high AERs in this method may have yielded unrealistic results due to two phenomena: (i) higher than normal air speeds through the enclosure assemblies (which may have altered mass transfer-limited reactions within the enclosure); and (ii) preferential airflow through leakage pathways that may not occur during periods of natural infiltration.

In this work, we developed an improved method for measuring ozone penetration factors in residences operating under natural infiltration conditions without artificial depressurization using a new NO-scrubbed ozone monitor that has lower detection limits and is less prone to interference. We applied the method in an unoccupied and sparsely furnished test apartment in Chicago, IL. Twenty-four repeated measurements of ozone penetration factors were made, providing a dataset for the following analyses: (i) an exploration of the accuracy and repeatability of the improved natural infiltration test method using multiple mathematical methods of solving for both ozone decay rate constants and penetration factors from the data; (ii) side-by-side comparisons between two ozone monitors during natural infiltration tests (an 'interference-free' and a conventional UV absorbance ozone monitor that was used in the previous work); and (iii) comparisons of results from natural infiltration experiments to those conducted with a blower door installed to test the validity of the original method.

Methods

Test apartment description

Measurements were conducted from June 2014 to August 2014 in *studioE* (the Suite for Testing Urban Dwellings and their Indoor and Outdoor Environments), an unoccupied and sparsely furnished

apartment unit on the third floor of Carman Hall on the main campus of Illinois Institute of Technology in Chicago, IL (Figures S1 and S2). The unit has a concrete floor covered with painted tiles and consists of two bedrooms, one living room, one bathroom, and a kitchen, with a floor area of ~60 m^2 and a volume of ~150 m^3. The interior walls are painted plaster. The exterior enclosure is made up of painted concrete block walls and single-pane aluminum-framed windows at about a 50:50 window-to-wall ratio. Only about half of the perimeter enclosure consists of exterior walls; the ceiling, floor, and other half of perimeter enclosures are all adjacent to other interior apartment and hallway spaces.

There is a central 100% recirculating air-handling unit that is connected to rigid sheet metal ductwork installed within conditioned space, but it is not connected to a heating or cooling system (it is only there to mimic a typical residential air handler and distribution system). A stand-alone air-conditioning unit provided cooling prior to measurements on hot days, although it was never operated during testing. The apartment unit typically receives ventilation air through infiltration through the exterior building enclosure and between the adjacent units, as well as through infiltration of supply air from the hallway through an undercut in the only doorway. However, the undercut was taped shut during measurements to limit intentional mechanical ventilation supply from the hallway. Two mechanical exhaust grilles within the unit were also taped shut during all measurements. All windows and the only door were kept closed during the measurements. The internal doors between each room were kept opened, and several oscillating fans were operated in the corner of each room to encourage mixing. No additional indoor sources of ozone were present during testing. The unit had no furnishings other than the measuring equipment and thus a low surface area to volume ratio relative to most furnished residences (Hodgson et al., 2005).

Measurements of ozone concentrations and air exchange rates

Indoor and outdoor ozone concentrations were measured using two UV absorbance ozone monitors: (i) a 2B Technologies Model 205 dual beam ozone monitor; and (ii) a 2B Technologies Model 211 'interference-free' scrubber-less ozone monitor, both connected to the same automated sampling system and operated at the same time with the same air sampling conditions. While both monitors are approved on US EPA's list of designated federal equivalent methods for measuring ambient ozone concentrations (U.S. EPA, 2014b), the monitors differ in their method of operation and detection limits, which has important implications for use in indoor environments. The Model 205 ozone monitor uses a conventional solid-phase scrubber and has a reported accuracy of 1 ppb and detection limit of 2 ppb. The 'interference-free' Model 211 monitor uses a NO-O_3 gas-phase titration scrubber in place of a solid-phase scrubber and has a reported accuracy of 1 ppb and detection limit of 1 ppb. N_2O was supplied by a small cylinder connected to a mass flow controller (Aalborg Model GFC17; Aalborg, Orangeburg, NY, USA) supplying N_2O at a rate of ~12 ml/min. N_2O is photolyzed to NO inside the monitor, which then reacts rapidly with O_3 to more selectively remove O_3 but not other potentially interfering compounds that absorb UV light in the same wavelength. We will continue to refer to the two ozone monitors as 'Model 205' and 'Model 211' for simplicity.

Both of the ozone monitors were placed in the living room and logged data at 10-s intervals. The ozone monitors were connected to a Swagelok Model SS-43GXS4-42DCX electrically actuated three-way ball valve to alternately monitor indoor and outdoor air. The valve was connected to an electronic timer (Sestos B3S-2R-24) and set to alternately measure indoor and outdoor ozone concentrations at 3-min intervals, alternating between 2-min indoors and 1-min outdoors. PTFE-lined sampling lines approximately 3 m in length were used for both indoor and outdoor sampling. Outdoor samples were drawn through a small (~0.6 cm) sampling line penetration in a clear acrylic window in the living room. Because the switching valve requires ~5 s to transition between indoor and outdoor sampling, the ozone monitors have a ~10–20 s response time after ozone concentrations change from indoors to outdoors (or vice versa), and it was not possible to always directly align the data logging starting time with the starting time of the switching valve, a few data points at both the beginning and end of each test period were dropped to make clear distinctions between indoor and outdoor sampling periods. Of 12 possible consecutive 10-s data points logged during each 2-min indoor sampling period, the middle nine data points were used to represent indoor ozone concentrations, with one or two data points typically shed on either side of the transition, depending on timing. These transition points were identified visually. Similarly, of the six possible consecutive 10-s data points logged during each 1-min outdoor sampling period, the middle three data points were used to represent outdoor ozone concentrations. The entire sampling system was well sealed, and the total ozone transport loss was <10% throughout the sampling lines and switching valve as assessed using periodic measurements with an ozone calibration source (2B Technologies Model 306; 2B Technologies, Boulder, CO, USA). Both ozone monitors were calibrated on a weekly basis using the same ozone calibration source. Transport loss was the same for indoor and outdoor samples from both systems.

Air exchange rates were measured during each test using CO_2 as a tracer gas. Periodic measurements of CO_2 concentrations inside the apartment unit

confirmed that there were no other significant CO_2 sources from adjacent apartments. An automatic CO_2 injection system was located in one of the bedrooms, which consisted of a small cylindrical CO_2 tank and an electronically powered solenoid valve regulator. At the beginning of each test, CO_2 was injected into the apartment for a period of approximately 15 min, typically yielding a peak concentration of ~1500 ppm inside. The subsequent decay was measured by two CO_2 monitors (PP Systems SBA-5; ±20 ppm accuracy) operated in the living room, both logging at 30-s intervals. One measured indoor CO_2 concentrations and the other measured outdoor CO_2 concentrations through another small sampling line penetration in another clear acrylic window. The two monitors were periodically co-located against each other, and linear regression coefficients were used to post-process raw CO_2 data into calibrated data. The AER was estimated by regressing the natural logarithm of the tracer gas concentrations vs. time, as shown in Equation (2) (ASTM E 741, 2006).

$$-\mathrm{In}\frac{Y_{\mathrm{in},t} - Y_{\mathrm{out}}}{Y_{\mathrm{in},t=0} - Y_{\mathrm{out}}} = \lambda t \qquad (2)$$

where $Y_{\mathrm{in},t}$ and $Y_{\mathrm{in},t\ =\ 0}$ are the indoor CO_2 concentrations (ppm) measured at time t and $t = 0$, respectively. Y_{out} is the average outdoor CO_2 concentration (ppm) during the test, and λ is the average AER (per hour). The tracer decay typically lasted the same duration of ozone penetration measurements (~3–4 h). Only data that clearly fit the log-linear exponential decay function in Equation (2) (with $R^2 > 0.99$) were used to estimate AERs. Data from periods of any drastic changes due to changing meteorological conditions were discarded. To test the extent of mixing, we performed initial tests in which we injected CO_2 in the middle of the living room and measured CO_2 concentrations using calibrated (via co-location) SBA-5 CO_2 monitors at six locations throughout the apartment unit. Differences in CO_2 concentrations between each location were <4%, and differences in response times were <10 s. Thus, the test apartment unit was considered to be reasonably well mixed.

Ozone penetration test method

Because the measurement of the ozone penetration factor (P) in a home also requires the knowledge of the simultaneous indoor ozone decay rate constant (k) in addition to λ from above, we explored a variety of methods to jointly estimate the two unknown parameters. We first conducted a preliminary experiment by alternately monitoring indoor and outdoor ozone concentrations over a period of 24 h with injection and decay of CO_2 every 4 h to measure the AER. This was similar to the method used by Rim et al. (2010) to estimate values of P and k for size-resolved particle infiltration by minimizing the sum of the absolute differences between observed indoor concentrations and those modeled using a discretized form of the mass balance in Equation (3) (Rim et al., 2010). However, this method proved to be too insensitive to solve for both P and k simultaneously because the natural indoor ozone concentration did not vary enough over this period to yield accurate estimates. Moreover, 24-h measurements in an actual occupied home would be impractical for widespread application.

Next, we refined our test procedure by combining techniques from our previous O_3 penetration test method (Stephens et al., 2012) and those of previous outdoor particle penetration measurements into homes (Chao et al., 2003; Stephens and Siegel, 2012a; Thatcher et al., 2003) into an improved method using a manual indoor ozone elevation and decay procedure during natural conditions. Indoor ozone concentrations were elevated using three UV ozone generators (CAP Model OZN-1, China) located in the bedrooms. The ozone generators photolyzed ambient indoor air filtered with activated carbon. We first used a wide variety of initial ozone concentrations ranging from 50 to 350 ppb to evaluate whether estimates of first-order decay rates were influenced by initial indoor ozone concentrations, but there was no correlation ($R^2 = 0.006$). Therefore, to achieve a clear exponential ozone decay profile while minimizing the test duration, the ozone generators were typically operated for approximately 12 min at the beginning of each test to elevate indoor ozone concentrations to ~120–160 ppb. AERs were measured simultaneously using CO_2 injection and decay.

Twenty-four experiments were conducted in the test apartment, including 21 repeated tests under natural conditions, six of which involved intentionally increasing k to test the sensitivity of P determinations to values of k, and three additional tests using the blower door method for comparison. To increase k in the six enhanced decay experiments, we installed activated carbon filtration in the return grille of the 100% recirculating central air-handling unit. In one of the six elevated k experiments, we also hung fabric curtains and distributed a variety of worn clothing materials throughout the unit to increase surface areas and introduce more reactive materials (Wisthaler and Weschler, 2010). Each experiment lasted 3–4 h and was typically conducted between 3 pm and 7 pm when outdoor ozone concentrations were highest.

Data analysis and parameter estimates

We used a dynamic mass balance approach to model the indoor ozone concentration in the well-mixed environment and in the absence of indoor sources

(Equation 3). To solve for the two unknown parameters in Equation (3) (P and k), we explored five solution methods: (i) a steady-state solution, (ii) a one-parameter analytical solution, (iii) a two-parameter analytical solution, (iv) a one-parameter discretized solution, and (v) a two-parameter discretized solution. In each case, the air exchange rate (AER, or λ) was estimated using Equation (2).

$$\frac{dC_{in}}{dt} = P\lambda C_{out} - (\lambda + k)C_{in} \quad (3)$$

In the steady-state method, P was estimated using time-averaged values of $C_{in,}$ and C_{out} during a steady-state period (Equation 4). The first-order ozone decay rate constant (k) was estimated using a log-linear regression solution to the initial portion of indoor decay that was not affected by outdoor ozone sources (Equation 5).

$$P = \frac{C_{in}}{C_{out}}\frac{\lambda + k}{\lambda} \quad (4)$$

$$-\ln\frac{C_{in}, t}{C_{in,t=0}} = (\lambda + k)\, t \quad (5)$$

where $C_{in,t}$ and $C_{in,t\ =\ 0}$ are the indoor ozone concentration at time t and $t = 0$, respectively. The initial log-linear portion was identified graphically by plotting the natural logarithm of indoor ozone concentration vs. time, which typically consisted of the first 10–30 min of data, depending on $C_{in,t\ =\ 0}$, λ, and the use of any means to increase k. We then estimated both λ and k during this period and used only the initial data that yielded $R^2 > 0.99$ to define the log-linear decay period. We should note that Equation 5 ignores background indoor ozone concentrations, which was reasonable given our elevated initial conditions. The mean values for initial and background indoor ozone concentrations were 131 ppb and 4 ppb, respectively, as measured with the Model 211 monitor. However, if indoor ozone concentrations cannot be elevated to such high levels above background, background concentrations should be accounted for in Equation 5.

The one-parameter and two-parameter analytical solutions both utilized a nonlinear regression to the analytical solution to Equation (3) as shown in Equation (6).

$$C_{in,t} = C_{in,t=0}e^{-(\lambda+k)\,t} + \frac{P\lambda\overline{C_{out}}}{\lambda + k}(1 - e^{-(\lambda+k)t}) \quad (6)$$

The one-parameter analytical solution utilized a nonlinear regression with Equation (6) to estimate P with prior knowledge of k from Equation (5). The two-parameter analytical solution utilized a nonlinear regression to estimate both P and k simultaneously, which is the same solution procedure that was used in our previous study (Stephens et al., 2012). Outdoor ozone concentrations were assumed to remain constant during each test period. However, this assumption was not always satisfied, which motivates the use of the last solution method below.

Because outdoor ozone concentrations were not always constant during the test periods, a discretized solution to Equation (3) was used to estimate parameters, as shown in Equation (7).

$$C_{in,t} = P\lambda C_{out,t}\Delta t + (1 - (\lambda + k)\Delta t)C_{in,t-1} \quad (7)$$

where $C_{in,t}$ and $C_{out,t}$ are the indoor and outdoor ozone concentrations at time t, respectively, and Δt is the time step (Δt = 3 min). In the one-parameter discretized solution, P was estimated using a nonlinear least-squares regression combined with the earlier estimate of k from Equation (5). This method is most similar to a method recently used to measure particle penetration through building enclosures with high accuracy (Stephens and Siegel, 2012). In the two-parameter discretized solution, both P and k were estimated simultaneously using a two-parameter nonlinear least-squares regression of the discretized solution in Equation (7).

Estimation of uncertainty

We considered a variety of approaches to estimate uncertainties associated with our estimates of P and k (Allen et al., 2006; Long et al., 2001; Rim et al., 2010; Stephens and Siegel, 2012a). First, the uncertainty in each AER estimate was calculated using the standard errors of the regression coefficients from Equation (2) and the average accuracy of the CO_2 monitors ($\pm$20 ppm) added in quadrature (Equation S1). Second, the uncertainty in each estimate of k was calculated by combining the standard error of the regression coefficient from Equation (5) with the AER uncertainties (Equation S2). Finally, the propagated uncertainty in P was estimated for each solution method by error propagation with a combination of relative standard deviations of average ozone concentration measurements (methods 1–3), relative uncertainties of k and AER (methods 1, 2, and 4), and the standard error of regression coefficients for P (methods 2–5), as shown in Equations S3 through S5. All parameter and uncertainty estimates were performed using a statistical software package, Stata version 12 (StataCorp SE, College Station, TX, USA).

Results and discussion

An example of alternating indoor and outdoor ozone concentration data from two consecutive ozone

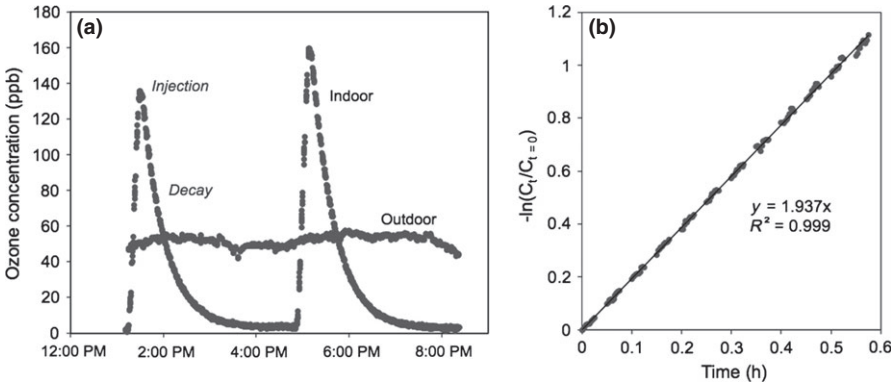

Fig. 1 Example of data from ozone injection and decay measurements using the Model 211 ozone monitor: (a) time-series data from two consecutive tests and (b) solving for the ozone decay rate constant using Equation (5)

injection and decay experiments on the same day is shown in Figure 1a (Model 211 data only). The data used to solve for the first-order decay rate constant (k) for one experiment is shown in Figure 1b. The AER (±propagated uncertainty) during this test was 0.19 ± 0.02 per hour, and the estimated value of k was 1.75 ± 0.02 per hour.

Ozone monitor comparison

Figure 2 summarizes steady-state indoor and outdoor ozone concentrations measured by both ozone monitors in the 21 replicate tests performed in the apartment unit under natural infiltration conditions (i.e., the last 10–30 min of each test similar to that shown in Figure 1a).

Both ozone monitors consistently yielded similar outdoor concentrations. The mean (±s.d.) outdoor concentration was 51.2 ± 12.9 ppb with the Model 205 monitor and 51.8 ± 12.5 ppb with the Model 211 monitor, both ranging from 23 to 70 ppb across all tests. Results from the two monitors were not statistically different from one another (P = 0.78 according to a Wilcoxon rank-sum test). However, the Model 205 monitor yielded significantly higher steady-state indoor ozone concentrations than the Model 211 monitor, with a mean (±s.d.) of 7.2 ± 2.4 ppb compared to 4.0 ± 1.3 ppb. These differences led to higher I/O ozone concentration ratios estimated with the Model 205, with a median of 0.14 compared to 0.09 (P = 0.0001 according to a Wilcoxon rank-sum test). These initial results demonstrate that the conventional UV absorbance ozone monitor (Model 205) tended to systematically overestimate indoor concentrations due to relatively high limits of detection and/or interference at low indoor ozone concentrations. This has important implications for using I/O ozone concentration data to estimate penetration factors under natural infiltration conditions. For example, data from the Model 205 monitor were used to solve for P using all five solution methods described in Equations (4) through (7), which yielded a mean (±s.d.) of ~1.07 ± 0.34 (results shown in Figure S3). Given that the penetration factor cannot theoretically exceed unity (Liu and Nazaroff, 2001), we chose to focus only on the Model 211 results in the remainder of this work to demonstrate the utility of the new natural infiltration test method.

Results from the estimates of λ, k, and P from all 21 natural ozone infiltration experiments using the Model 211 ozone monitor and all five solution methods are shown in Table S1, along with average outdoor ozone concentrations measured during each test. The mean

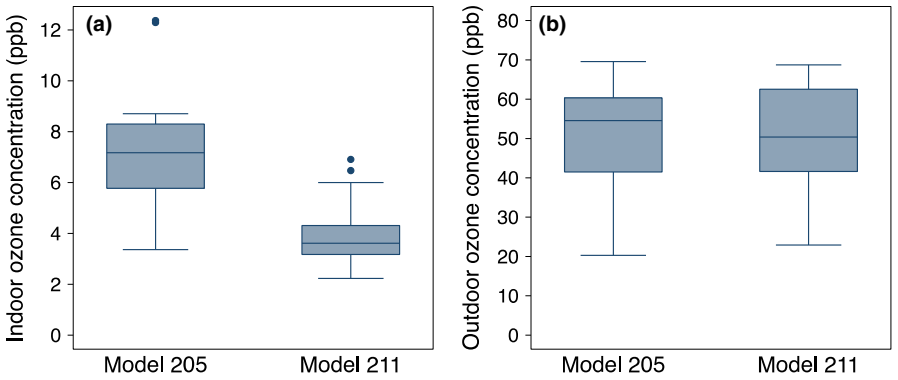

Fig. 2 Steady-state (a) indoor and (b) outdoor ozone concentrations measured across 21 replicate ozone penetration tests by two ozone monitors

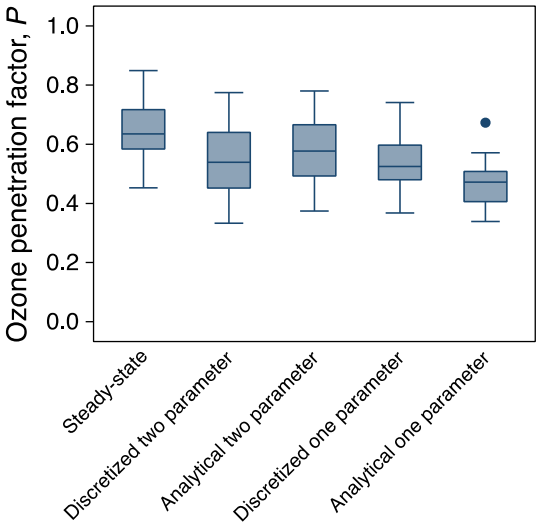

Fig. 3 Estimates of ozone penetration factors (P) across 21 replicate experiments under natural infiltration conditions using five solution methods and the Model 211 ozone monitor. Values of k ranged 1.2–3.3 per hour depending on indoor surface area characteristics

estimates (±s.d.) of P over all 21 natural infiltration tests in the test apartment were 0.65 ± 0.11, 0.47 ± 0.08, 0.59 ± 0.11, 0.54 ± 0.10, and 0.55 ± 0.12 using the steady-state, one-parameter analytical, two-parameter analytical, one-parameter discretized, and two-parameter discretized solutions, respectively (shown in Figure 3). Differences in each solution method are further explored in the next section.

Best estimates of k ranged from 1.24 to 3.30 per hour depending on indoor surface area characteristics. In the 15 experiments in which we did not attempt to artificially increase k, the mean (±s.d.) estimate of k was 1.62 ± 0.18 per hour (estimated using Equation 5). In the six experiments in which we attempted to artificially increase k, the mean (±s.d.) estimate of k was increased to 2.51 ± 0.43 per hour. The highest value occurred during an experiment conducted with a combination of activated carbon sheets installed in the return grille of the air-handling unit, fabric curtains over the windows, and worn clothing distributed throughout the apartment unit (~3.28 per hour on September 29, 2014). Estimates of k under these conditions were closer to that which has been observed in occupied residences under normal conditions (Lee et al., 1999; Weschler, 2000).

Comparison of solution methods

Estimates of P were reasonably similar across all solution methods, particularly for the two discretized solution methods and the two-parameter analytical solution method. Median estimates of P ranged from a maximum of 0.64 using the steady-state solution method to 0.48 using the analytical one-parameter solution. Estimates of P using the steady-state method were significantly higher than the other solution methods ($P < 0.05$ for all comparisons but the analytical two-parameter method using a Wilcoxon rank-sum test). Conversely, estimates of P made using the analytical one-parameter method were significantly lower than all other solution methods (Wilcoxon rank-sum $P < 0.05$). There were no significant differences in estimates of P between the discretized one-parameter, discretized two-parameter, or the analytical two-parameter solution.

Two metrics were used to further estimate model fit and accuracy of each solution method using the Model 211 monitor data: (i) uncertainty estimates; and (ii) mean squared errors (MSE). The steady-state method had the highest average (±s.d.) uncertainty estimate at 0.19 ± 0.05, or 29 ± 5% on a relative basis, as shown in Figure 4a. High uncertainty stems largely from the reliance on large relative standard deviations of the

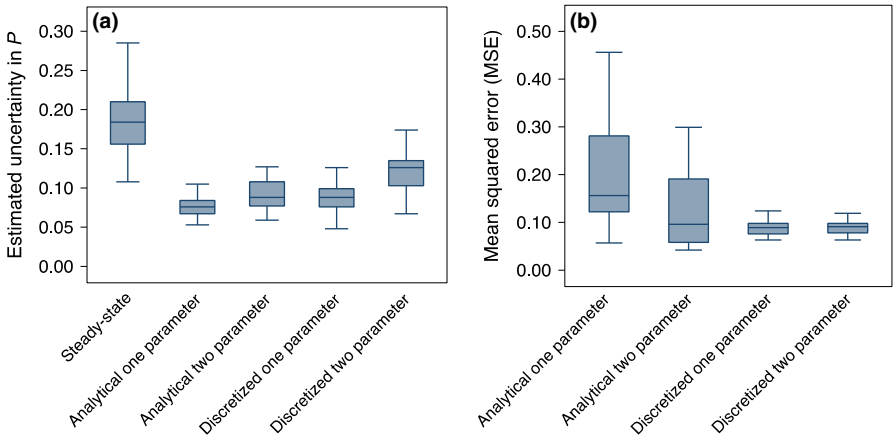

Fig. 4 Range of uncertainty estimates in 21 natural infiltration experiments using the Model 211 ozone monitor: (a) estimated uncertainty in ozone penetration factor estimated by all five solution methods and (b) mean squared error (MSE) for the analytical and discretized solution methods

Table 1 Summary of estimates of λ, k, and P (using the one-parameter discretized solution method) and indoor and outdoor environmental conditions

Date	P (Equation 7) (–)	k (Equation 5) (per hour)	AER, λ (Equation 2) (per hour)	Outdoor ozone (ppb)	Wind dir. (from N)	Wind speed (m/s)	Outdoor temp. (°C)	Outdoor RH (%)	Indoor temp. (°C)	Indoor RH (%)
June 17, 2014	0.37 ± 0.04	1.69 ± 0.04	0.41 ± 0.04	68 ± 3	225	0.67	32.3	42	34.5	46
June 27, 2014	0.41 ± 0.03	1.56 ± 0.03	0.24 ± 0.02	62 ± 3	157.5	1.49	31.1	44	32.2	46
July 9, 2014	0.49 ± 0.04	1.92 ± 0.04	0.32 ± 0.03	31 ± 1	90	0.56	21.7	68	29.5	51
July 15, 2014	0.74 ± 0.06	2.49 ± 0.06	0.55 ± 0.06	23 ± 1	315	0.67	18.9	66	27.9	44
July 17, 2014	0.63 ± 0.02	1.77 ± 0.02	0.19 ± 0.02	51 ± 2	112.5	0.71	23.1	53	28.7	43
July 18, 2014	0.68 ± 0.02	1.78 ± 0.02	0.19 ± 0.02	51 ± 3	135	1.07	25.5	45	33.0	40
July 18, 2014	0.48 ± 0.02	1.75 ± 0.02	0.19 ± 0.02	53 ± 2	112.5	0.52	24.3	49	30.9	38
July 19, 2014	0.52 ± 0.02	2.61 ± 0.02	0.19 ± 0.02	67 ± 3	135	0.36	24.6	49	33.5	41
July 22, 2014	0.69 ± 0.02	1.79 ± 0.02	0.19 ± 0.02	55 ± 3	202.5	1.41	32.0	53	33.8	49
July 25, 2014	0.60 ± 0.03	1.54 ± 0.03	0.22 ± 0.02	42 ± 3	225	1.28	23.9	53	29.1	43
July 25, 2014	0.61 ± 0.02	1.61 ± 0.02	0.18 ± 0.02	42 ± 3	225	0.67	23.5	58	30.4	43
July 31, 2014	0.52 ± 0.03	1.53 ± 0.03	0.30 ± 0.02	47 ± 2	270	0.72	29.3	35	29.4	46
July 31, 2014	0.53 ± 0.03	1.55 ± 0.03	0.24 ± 0.03	40 ± 4	270	0.30	27.8	39	30.9	44
August 5, 2014	0.54 ± 0.02	2.35 ± 0.02	0.22 ± 0.02	51 ± 6	45	1.09	23.2	77	30.6	58
August 7, 2014	0.44 ± 0.02	2.11 ± 0.02	0.19 ± 0.02	70 ± 3	45	0.85	25.2	56	29.0	56
August 7, 2014	0.58 ± 0.02	2.21 ± 0.02	0.17 ± 0.02	55 ± 4	45	1.44	23.4	69	30.6	53
August 10, 2014	0.50 ± 0.03	1.33 ± 0.03	0.24 ± 0.03	60 ± 2	45	0.92	24.9	61	29.3	54
August 10, 2014	0.56 ± 0.03	1.24 ± 0.03	0.26 ± 0.02	61 ± 5	45	0.59	23.9	66	28.1	56
August 22, 2014	0.40 ± 0.05	1.75 ± 0.05	0.47 ± 0.05	65 ± 3	90	0.23	29.2	68	32.0	64
August 26, 2014	0.47 ± 0.03	1.58 ± 0.03	0.29 ± 0.03	42 ± 2	90	0.14	26.9	65	31.2	57
September 29, 2014	0.59 ± 0.03	3.30 ± 0.03	0.16 ± 0.02	62 ± 2	315	0.22	27.7	34	28.3	51
Mean	0.54	1.88	0.26	52	152	0.76	25.8	55	30.6	49
s.d.	0.10	0.49	0.10	12	93	0.41	3.5	12	2.0	7

steady-state indoor and outdoor ozone concentrations used in this solution method. The next highest average uncertainty was estimated with the two-parameter discretized solution method (0.13 ± 0.04). The average (±s.d.) uncertainties of the one- and two-parameter analytical and one-parameter discretized solutions were similar: 0.08 ± 0.01, 0.09 ± 0.02, and 0.09 ± 0.03, respectively. These data suggest that either of the two analytical or the one-parameter discretized solution methods is most appropriate to use for minimizing uncertainty.

As a measure of model fit between the four dynamic solution methods, the MSE was calculated for all 21 natural infiltration experiments using data from both of the analytical and discretized solution methods, as shown in Figure 4b. The MSEs of the two analytical solution methods were higher than the MSEs of the two discretized solution methods, which were very similar. The average (±s.d.) MSE of the one- and two-parameter analytical solution methods were 0.26 ± 0.23 and 0.15 ± 0.16, respectively, compared to only 0.10 ± 0.04 for both the one- and two-parameter discretized solution methods. Thus, both discretized solution methods achieve a low MSE, while the discretized one-parameter method also yields the lowest estimated uncertainty.

Overall, these data suggest that measurements with the Model 211 ozone monitor combined with parameter estimates made using the one-parameter discretized solution method yield the most repeatable estimates of both P and k under natural infiltration conditions with the lowest combined uncertainty and MSE. This is most likely because the discretized solution method takes into account varying outdoor concentrations, while the use of data from the initial decay period yields reliable estimates of k. Differences in the ability of the analytical and discretized solution methods to accurately predict indoor ozone concentrations are further explored in Figure S3, which plots the inverse of modeled indoor ozone concentrations vs. the inverse of measured indoor ozone concentrations to evaluate model predictions at very low indoor ozone concentrations achieved at the end of each test. A full summary of estimates of uncertainty in P, MSEs, and R^2 values for these model fits is also provided in Table S2.

Influence of environmental conditions

Results for P and k during the 21 natural infiltration tests estimated using the one-parameter discretized solution were then used to explore correlations between several potentially influential indoor and outdoor environmental factors (e.g., averages of indoor/outdoor temperature and relative humidity, outdoor ozone concentrations, and wind speed and direction during the tests) (Table 1). Data for outdoor temperature, relative humidity, and wind speeds and directions were taken at 5-min intervals from a publicly accessible weather station at US Cellular Field approximately 1 km from the apartment unit (Weather Underground, 2014). Indoor temperature and relative humidity were measured using an Onset HOBO U12 logging at 5-min

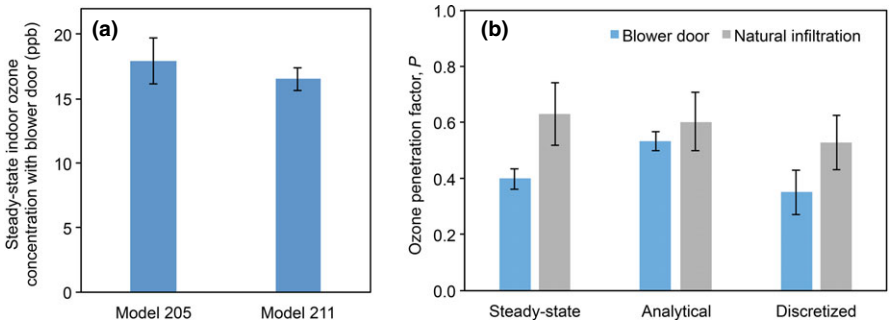

Fig. 5 Comparison of natural infiltration results to the blower door test method: (a) steady-state indoor ozone concentrations using both ozone monitors with the blower door operating during three replicate tests and (b) comparison of estimates of ozone penetration factor (P), averaged over three blower door tests and 21 natural infiltration tests

intervals. The average indoor and outdoor temperatures were 30.6°C (range 27.9–34.5°C) and 25.8°C (range 18.9–32.3°C), respectively. The average indoor humidity and outdoor relative humidity were 49% (range 38–64%) and 55% (range 34–77%), respectively. Spearman's rank correlations between P, k, λ, and all environmental conditions reported in Table 1 did not reveal any significant relationships between any of these parameters ($P > 0.05$), suggesting that the test and solution methods are not measurably affected by these environmental conditions.

Comparison to the original blower door test method

Finally, a subset of three tests was performed using the original fan depressurization test method reported in Stephens et al. (2012). A blower door was placed in the doorway, and the unit was depressurized with respect to the hallway, elevating λ to ~3 per hour. Both ozone monitors were used for these tests for comparison. Figure 5a shows the steady-state indoor ozone concentrations measured using data from both ozone monitors. The mean (±s.d.) indoor concentrations were similar: 16.5 ± 0.9 ppb and 17.9 ± 1.8 ppb with the Model 211 and 205 monitors, respectively ($P = 0.28$ according to a Wilcoxon signed-rank test), suggesting that the fan depressurization method was able to elevate indoor ozone concentrations above the lower limit of detection of the Model 205 monitor.

Parameter estimates for λ, k, and P during the three blower door experiments were made using three solution methods (steady-state, two-parameter analytical, and one-parameter discretized) and compared to the mean (±s.d.) from the 21 natural infiltration experiments in Figure 5b. These data suggest that in this building, the use of the blower door method actually underestimated the ozone penetration factor relative to that measured during natural infiltration for all three solution methods, although the two-parameter analytical solution agreed best, suggesting that previous measurements of P in Stephens et al. (2012) may still be considered reasonable estimates.

These data are the first known measurements of ozone penetration factors for a residential building or multifamily apartment unit operating under natural infiltration conditions. The mean (±s.d.) estimate of the envelope penetration factor (P) using this improved method with a one-parameter discretized solution was 0.54 ± 0.10 across a wide range of indoor and outdoor environmental conditions. Estimates of P ranged from 0.37 ± 0.05 to 0.74 ± 0.12, with most of the data relatively tightly grouped within 20% of the mean value, although some days had deviations as high as 32–37% from the mean. Results from this unoccupied and sparsely furnished apartment suggest that the ozone penetration factor is lower than what is typically assumed in most residences, which has important implications for the magnitude and type of ozone reaction byproducts found indoors. Given the repeatability and accuracy of the improved test method, we recommend that it be used to measure ozone penetration factors in a greater number and variety of building types to improve our knowledge of indoor exposures to outdoor ozone and ozone reaction byproducts.

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Supporting Information

Additional Supporting Information may be found in the online version of this article:
Figure S1. *StudioE*: The Suite for Testing Urban Dwellings and their Indoor and Outdoor Environments, the unoccupied apartment unit used in this work.

Figure S2. Ozone instrumentation setup and floor plan in the test apartment unit *StudioE*.
Figure S3. Estimates of ozone penetration factors (P) across 21 replicate experiments under natural infiltration conditions using five solution methods and 2B Technologies Model 205 ozone monitor.
Figure S4. Example of model fits for (a) the one-parameter discretized solution method and (b) the two-parameter analytical solution method.

Table S1. Summary of results from five solution methods during 21 natural infiltration experiments with the Model 211 ozone monitor.
Table S2. Summary of estimates of uncertainties (*Unc*.), mean squared errors (MSE), and R^2 values (R^2) using the five solution methods.

References

Allen, R., Wallace, L., Larson, T., Sheppard, L. and Liu, L.-J.S. (2006) Evaluation of the recursive model approach for estimating particulate matter infiltration efficiencies using continuous light scattering data, *J. Expo. Sci. Environ. Epidemiol.*, **17**, 468–477.

ASTM D5156 (2008) Standard Test Methods for Continuous Measurement of Ozone in Ambient, Workplace, and Indoor Atmospheres (Ultraviolet Absorption).

ASTM E 741 (2006) Standard Test Method for Determining Air Change in a Single Zone by Means of a Tracer Gas Dilution.

Avol, E.L., Navidi, W.C. and Colome, S.D. (1998) Modeling ozone levels in and around southern California homes, *Environ. Sci. Technol.*, **32**, 463–468.

Bell, M., McDermott, A., Zeger, S., Samet, J. and Dominici, F. (2004) Ozone and short-term mortality in 95 US urban communities, *J. Am. Med. Assoc.*, **292**, 2372–2378.

Bell, M.L., Peng, R.D. and Dominici, F. (2006) The exposure-response curve for ozone and risk of mortality and the adequacy of current ozone regulations, *Environ. Health Perspect.*, **114**, 532–536.

Bell, M.L., Zanobetti, A. and Dominici, F. (2014) Who is more affected by ozone pollution? A systematic review and meta-analysis, *Am. J. Epidemiol.*, **180**, 15–28.

Brauner, E.V., Karottki, D.G., Frederiksen, M., Kolarik, B., Spilak, M., Andersen, Z.J., Vibenholt, A., Ellermann, T., Gunnarsen, L. and Loft, S. (2014) Residential ozone and lung function in the elderly, *Indoor Built Environ*. Available from: http://ibe.sagepub.com/cgi/doi/10.1177/1420326X14539339 (accessed August 21, 2014).

Cattaneo, A., Peruzzo, C., Garramone, G., Urso, P., Ruggeri, R., Carrer, P. and Cavallo, D.M. (2011) Airborne particulate matter and gaseous air pollutants in residential structures in Lodi province, Italy, *Indoor Air*, **21**, 489–500.

Chan, W.R., Nazaroff, W.W., Price, P.N., Sohn, M.D. and Gadgil, A.J. (2005) Analyzing a database of residential air leakage in the United States, *Atmos. Environ.*, **39**, 3445–3455.

Chao, C.Y.H., Wan, M.P. and Cheng, E.C.K. (2003) Penetration coefficient and deposition rate as a function of particle size in non-smoking naturally ventilated residences, *Atmos. Environ.*, **37**, 4233–4241.

Chen, C., Zhao, B. and Weschler, C.J. (2011) Assessing the influence of indoor exposure to 'outdoor ozone' on the relationship between ozone and short-term mortality in U.S. communities, *Environ. Health Perspect*. Available from: http://www.ehponline.org/ambra-doi-resolver/10.1289/ehp.1103970 (accessed November 19, 2011).

Fadeyi, M.O., Weschler, C.J., Tham, K.W., Wu, W.Y. and Sultan, Z.M. (2013) Impact of human presence on secondary organic aerosols derived from ozone-initiated chemistry in a simulated office environment, *Environ. Sci. Technol.*, **47**, 3933–3941.

Fann, N., Lamson, A.D., Anenberg, S.C., Wesson, K., Risley, D. and Hubbell, B.J. (2012) Estimating the national public health burden associated with exposure to ambient PM2.5 and ozone, *Risk Anal.*, **32**, 81–95.

Gall, E.T., Corsi, R.L. and Siegel, J.A. (2011) Barriers and opportunities for passive removal of indoor ozone, *Atmos. Environ.*, **45**, 3338–3341.

Georgopoulos, P.G., Wang, S.-W., Vyas, V.M., Sun, Q., Burke, J., Vedantham, R., McCurdy, T. and Özkaynak, H. (2005) A source-to-dose assessment of population exposures to fine PM and ozone in Philadelphia, PA, during a summer 1999 episode, *J. Expo. Anal. Environ. Epidemiol.*, **15**, 439–457.

Grosjean, D. and Harrison, J. (1985) Response of chemiluminescence NOx analyzers and ultraviolet ozone analyzers to organic air pollutants, *Environ. Sci. Technol.*, **19**, 862–865.

Hodgson, A.T., Ming, K.Y. and Singer, B.C. (2005) *Quantifying Object and Material Surface Areas in Residences*, Berkeley, CA, Lawrence Berkeley National Laboratory.

Huntzicker, J.J. and Johnson, R.L. (1979) Investigation of an ambient interference in the measurement of ozone by ultraviolet absorption photometry, *Environ. Sci. Technol.*, **13**, 1414–1416.

Ito, K., Leon, S.F.D. and Lippmann, M. (2005) Associations between ozone and daily mortality: analysis and meta-analysis, *Epidemiology*, **16**, 446–457.

Jerrett, M., Burnett, R.T., Pope, C.A. 3rd, Ito, K., Thurston, G., Krewski, D., Shi, Y., Calle, E. and Thun, M. (2009) Long-term ozone exposure and mortality, *N. Engl. J. Med.*, **360**, 1085–1095.

Johnson, T., Capel, J. and Ollison, W. (2014) Measurement of microenvironmental ozone concentrations in Durham. North Carolina, using a 2B Technologies 205 FEM monitor and an interference-free 2B Technologies 211 monitor, *J. Air Waste Manag. Assoc.*, **64**, 360–371.

Klepeis, N.E., Nelson, W.C., Ott, W.R., Robinson, J.P., Tsang, A.M., Switzer, P., Behar, J.V., Hern, S.C. and Engelmann, W.H. (2001) The National Human Activity Pattern Survey (NHAPS): a resource for assessing exposure to environmental pollutants, *J. Expo. Anal. Environ. Epidemiol.*, **11**, 231–252.

Kunkel, D.A., Gall, E.T., Siegel, J.A., Novoselac, A., Morrison, G.C. and Corsi, R.L. (2010) Passive reduction of human exposure to indoor ozone, *Build. Environ.*, **45**, 445–452.

Lee, K., Vallarino, J., Dumyahn, T., Ozkaynak, H. and Spengler, J.D. (1999) Ozone decay rates in residences, *J. Air Waste Manag. Assoc.*, **49**, 1238–1244.

Lee, K., Parkhurst, W.J., Xue, J., Ozkaynak, A.H., Neuberg, D. and Spengler, J.D. (2004) Outdoor/indoor/personal ozone exposures of children in Nashville, Tennessee, *J. Air Waste Manag. Assoc.*, **54**, 352–359.

Liu, D. and Nazaroff, W.W. (2001) Modeling pollutant penetration across building envelopes, *Atmos. Environ.*, **35**, 4451–4462.

Liu, L.J., Koutrakis, P., Leech, J. and Broder, I. (1995) Assessment of ozone exposures in the greater metropolitan Toronto area, *J. Air Waste Manag. Assoc.*, **45**, 223–234.

Long, C.M., Suh, H.H., Catalano, P.J. and Koutrakis, P. (2001) Using time- and

size-resolved particulate data to quantify indoor penetration and deposition behavior, *Environ. Sci. Technol.*, **35**, 2089–2099.

Ollison, W.M., Crow, W. and Spicer, C.W. (2013) Field testing of new-technology ambient air ozone monitors, *J. Air Waste Manag. Assoc.*, **63**, 855–863.

Rim, D., Wallace, L. and Persily, A. (2010) Infiltration of outdoor ultrafine particles into a test house, *Environ. Sci. Technol.*, **44**, 5908–5913.

Romieu, I., Lugo, M.C., Colome, S., Garcia, A.M., Avila, M.H., Geyh, A., Velasco, S.R. and Rendon, E.P. (1998) Evaluation of indoor ozone concentration and predictors of indoor-outdoor ratio in Mexico City, *J. Air Waste Manag. Assoc.*, **48**, 327–335.

Shu, S. and Morrison, G.C. (2011) Surface reaction rate and probability of ozone and alpha-terpineol on glass, polyvinyl chloride, and latex paint surfaces, *Environ. Sci. Technol.*, **45**, 4285–4292.

Smith, R.L., Xu, B. and Switzer, P. (2009) Reassessing the relationship between ozone and short-term mortality in U.S. urban communities, *Inhal. Toxicol.*, **21**, 37–61.

Spicer, C.W., Joseph, D.W. and Ollison, W.M. (2010) A re-examination of ambient air ozone monitor interferences, *J. Air Waste Manag. Assoc.*, **60**, 1353–1364.

Stephens, B. and Siegel, J.A. (2012) Penetration of ambient submicron particles into single-family residences and associations with building characteristics, *Indoor Air*, **22**, 501–513.

Stephens, B., Gall, E.T. and Siegel, J.A. (2012) Measuring the penetration of ambient ozone into residential buildings, *Environ. Sci. Technol.*, **46**, 929–936.

Thatcher, T.L., Lunden, M.M., Revzan, K.L., Sextro, R.G. and Brown, N.J. (2003) A concentration rebound method for measuring particle penetration and deposition in the indoor environment, *Aerosol Sci. Technol.*, **37**, 847–864.

U.S. EPA (2007) *Ozone Population Exposure Analysis for Selected Urban Areas*, Research Triangle Park, North Carolina, U.S. Environmental Protection Agency.

U.S. EPA (2014a) *Health Risk and Exposure Assessment for Ozone: Final Report*, Research Triangle Park, North Carolina, U.S Environmental Protection Agency.

U.S. EPA (2014b) List of Designated Reference and Equivalent Methods, United States Environmental Protection Agency National Exposure Research Laboratory, Available from: http://www.epa.gov/ttnamti1/files/ambient/criteria/reference-equivalent-methods-list.pdf.

Walker, I.S. and Sherman, M.H. (2013) Effect of ventilation strategies on residential ozone levels, *Build. Environ.*, **59**, 456–465.

Wang, C. and Waring, M.S. (2014) Secondary organic aerosol formation initiated from reactions between ozone and surface-sorbed squalene, *Atmos. Environ.*, **84**, 222–229.

Waring, M.S. (2014) Secondary organic aerosol in residences: predicting its fraction of fine particle mass and determinants of formation strength, *Indoor Air*, **24**, 376–389.

Waring, M.S. and Siegel, J.A. (2013) Indoor secondary organic aerosol formation initiated from reactions between ozone and surface-sorbed D-limonene, *Environ. Sci. Technol.*, **47**, 6341–6348.

Waring, M.S. and Wells, J.R. (2014) Volatile organic compound conversion by ozone, hydroxyl radicals, and nitrate radicals in residential indoor air: magnitudes and impacts of oxidant sources, *Atmos. Environ.*, Available from: http://linkinghub.elsevier.com/retrieve/pii/S1352231014005172 (accessed August 21, 2014).

Weather Underground (2014) PWS Data: U.S. Cellular Field KILCHICA30, Personal Weather Station Network, Available from: http://www.wunderground.com/personal-weather-station/dashboard?ID=KILCHICA30.

Weschler, C.J. (2000) Ozone in indoor environments: concentration and chemistry, *Indoor Air*, **10**, 269–288.

Weschler, C.J. (2006) Ozone's impact on public health: contributions from indoor exposures to ozone and products of ozone-initiated chemistry, *Environ. Health Perspect.*, **114**, 1489–1496.

Williams, E.J., Fehsenfeld, F.C., Jobson, B.T., Kuster, W.C., Goldan, P.D., Stutz, J. and McClenny, W.A. (2006) Comparison of ultraviolet absorbance, chemiluminescence, and DOAS instruments for ambient ozone monitoring, *Environ. Sci. Technol.*, **40**, 5755–5762.

Wilson, K.L. and Birks, J.W. (2006) Mechanism and elimination of a water vapor interference in the measurement of ozone by UV absorbance, *Environ. Sci. Technol.*, **40**, 6361–6367.

Wisthaler, A. and Weschler, C.J. (2010) Reactions of ozone with human skin lipids: sources of carbonyls, dicarbonyls, and hydroxycarbonyls in indoor air, *Proc. Natl Acad. Sci. USA*, **107**, 6568–6575.

Zhang, J. and Lioy, P.J. (1994) Ozone in residential air: concentrations I/O ratios, indoor chemistry, and exposures, *Indoor Air*, **4**, 95–105.