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# **Quantifying Fine Particle Emission Events from Time-Resolved Measurements: Method Description and Application to 18 California Low-Income Apartments**

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## Abstract

PM<sub>2.5</sub> exposure is associated with significant health risk. Exposures in homes derive from both outdoor and indoor sources, with emissions occurring primarily in discrete events. Data on emission event magnitudes and schedules are needed to support simulation-based studies of exposures and mitigations. This study applied an identification and characterization algorithm to quantify time-resolved PM<sub>2.5</sub> emission events from data collected during 224 days of monitoring in 18 California apartments with low-income residents. We identified and characterized 836 distinct events with median and mean values of 12 and 30 mg emitted mass, 16 and 23 minutes emission duration, 37 and 103 mg/h emission rates, and pseudo-first order decay rates of 1.3 and 2.0/h. Mean event-averaged concentrations calculated using the determined event characteristics agreed to within 6% of measured values for 14 of the apartments. There were variations in event schedules and emitted mass across homes, with few events overnight and most emissions occurring during late afternoons and evenings. Event characteristics were similar during weekdays and weekends. Emitted mass was positively correlated with number of residents (Spearman coefficient,  $\rho=0.10$ ), bedrooms ( $\rho=0.08$ ), house volume ( $\rho=0.29$ ), and indoor-outdoor CO<sub>2</sub> difference ( $\rho=0.27$ ). The event schedules can be used in probabilistic modeling of PM<sub>2.5</sub> in low-income apartments.

**Key Words:** PM<sub>2.5</sub>, residential, exposure, multifamily, cooking

**Practical Implications:** The indoor emission events quantified in this study provide diurnal and weekly profiles that can be used to simulate PM<sub>2.5</sub> concentrations and exposures in low-income apartments. The algorithm can be used to determine emission event characteristics and profiles for other types of homes for which time-resolved indoor and outdoor PM<sub>2.5</sub> data are available.

## 1 Introduction

The U.S. Environmental Protection Agency's most recent Integrated Scientific Assessment for Particulate Matter<sup>1</sup> determined that both short-term and long-term exposures to elevated ambient PM<sub>2.5</sub> cause increases in cardiovascular effects and mortality; robustly established associations with respiratory effects were assessed as likely to be causal. Several recent studies have investigated health outcomes in relation to indoor particle exposures<sup>2-6</sup>.

The home environment is an important location of PM<sub>2.5</sub> intake, owing both to the amount of time spent at home<sup>7</sup> and the high intake fraction for indoor PM<sub>2.5</sub> sources<sup>8</sup>. The relative contributions of ambient and indoor-generated PM<sub>2.5</sub> vary widely across homes<sup>9-14</sup>. Using concentration–response functions derived

from analyses connecting ambient PM<sub>2.5</sub> to health effects, Logue et al.<sup>15</sup> found that intake of PM<sub>2.5</sub> is likely responsible for more chronic health impacts – measured in disability adjusted life years – than any other non-biological air pollutant in US residences.

PM<sub>2.5</sub> is emitted inside homes during events and activities including tobacco smoking, cooking and cooking burner use, burning of incense and candles, secondary aerosol formation from ozone-terpene reactions, and re-suspension, among other sources<sup>16-28</sup>. Numerous studies have reported elevated concentrations occurring during scripted or natural indoor activities in homes<sup>14, 17, 20, 27, 29-32</sup>.

Exposure to PM<sub>2.5</sub> in homes can be reduced by limiting source activities, managing ventilation and/or filtering the outdoor air supply, using kitchen exhaust ventilation to remove cooking-related particles<sup>33</sup>, and filtering indoor air via a forced air system or standalone devices<sup>34-38</sup>. PM<sub>2.5</sub> exposures and the effectiveness of controls can be investigated through measurements in intervention studies or under controlled conditions, or by applying simulation-based analysis to individual buildings or the building stock<sup>34-41</sup>. Simulation offers advantages over experimental or empirical evaluation, including the ability to investigate many more control alternatives under varying conditions.

Simulating residential PM<sub>2.5</sub> concentrations requires quantitative and time-resolved introduction of material from both outdoors and indoor emission sources. One approach to simulating indoor sources is to apply emission factors and frequency profiles for specific activities. Emission rates and emission factors for many sources have been reported from experiments conducted in laboratory test chambers or residences under controlled conditions. Emissions are calculated from the time concentration profile and knowledge of the air exchange rate, with the assumption of a well-mixed airspace<sup>16, 18, 25, 28</sup>. A few studies have analyzed time-resolved particle measurements and occupant diary data to associate identified indoor peaks with specific occupant activities<sup>19-21, 42</sup>. However, modeling of PM<sub>2.5</sub> emissions by probabilistically accounting for all relevant sources is still limited by the lack of data on source frequencies and schedules. An alternative approach is to use empirical emission event schedules developed from analysis of PM<sub>2.5</sub> data collected inside and outside of occupied homes.

The primary objectives of the present study were to demonstrate an algorithm-based technique to identify indoor emission events and quantify their characteristics from time-resolved particle data, and to apply the technique to determine event characteristics for data collected in 18 California apartments with low-income residents. The secondary objective was to investigate relationships between emissions and household characteristics for these homes. The ultimate goal of this effort is to construct a database of indoor particle emission events that can be used in data-driven, probabilistic modeling of PM<sub>2.5</sub> concentrations in homes.

## 2 Methods and Materials

### 2.1 Method to Identify and Characterize PM<sub>2.5</sub> Events

The characterization algorithm is based on an idealized model of a PM<sub>2.5</sub> emission event consisting of an emission period followed by decay. The model treats the source as constantly emitting into a completely and instantaneously well-mixed volume that is not simultaneously impacted by other indoor emission sources but has a baseline indoor concentration of particles from outdoors. The model treats each sharp increase above the baseline and subsequent decrease as a distinct event. All loss mechanisms are approximated as a pseudo-first order decay process. These are simplifications since actual PM<sub>2.5</sub> emission events may have time-varying emission rates, coincident sources, non-ideal mixing, and combinations of loss mechanisms that do not follow first order dynamics throughout the decay.

To characterize an event, we identify the beginning and end of the emission period, the peak concentration, and a suitable interval of decay to calculate a loss rate that is assumed to pertain throughout the emission event. A smoothed time series of the indoor PM<sub>2.5</sub> concentration measured during these periods is fitted to equations representing the idealized model to determine the pseudo-first order decay rate and emission rate for the event.

#### Overview of Approach

The algorithm includes the following steps, described in detail in the subsections that follow:

1. Start with time-resolved, indoor and outdoor particle concentration data. For this study we used 2-minute data from a study of 18 low-income apartments in California.
2. Calculate baseline indoor concentration of outdoor particles using building-specific infiltration factor and time-resolved outdoor particle data.
3. Smooth indoor data and identify peaks using an analysis package developed for chromatograms.
4. Identify the start time for each emission event and the end time for each decay period.
5. Identify linked events for which the decay of the earlier event corresponds to the start of emissions for the later event.
6. Visually review and add peaks that were missed by automatic algorithm. Repeat steps 4-6.
7. Determine pseudo-first order decay rate for each distinct event.
8. Calculate emission rate for each distinct event and for all events in series of linked events.

## **Step 1. Data Source: California Low-Income Apartment Study**

This study analyzed data from an evaluation of synergistic energy and indoor air quality retrofits in 18 California apartments with low-income residents<sup>43</sup>. Selection criteria for the study included: 1) subsidized housing for low-income residents, 2) apartment-level heating equipment, 3) no smoking allowed in home, and 4) informed consent signed by tenants and participation agreement signed by building owners. Table 1 presents summary characteristics for the apartments, which were spread equally among three complexes. All homes had gas heating and complexes 1 and 3 had air conditioning. Air quality parameters were measured at a central location in each apartment – typically the living room or dining room – over two weeks pre-retrofit and another two weeks post-retrofit. For this study only the pre-retrofit data were analyzed so that the results reflect indoor emission events in low-income apartments without designed controls; several retrofits, including standalone air filter units and commissioning or replacing venting range hoods to ensure adequate exhaust airflow, may have reduced the apparent PM<sub>2.5</sub> emission rates or accelerated particle removal following an event.

The apartment study<sup>44</sup> used light scattering monitors (DustTrak II 8530, TSI Inc., Shoreview, MN, USA) to obtain a surrogate measure of PM<sub>2.5</sub> in 2-minute intervals. DustTraks have been shown to overestimate ambient and indoor PM<sub>2.5</sub> concentrations by a factor of 1.9 to 5.6 in other studies<sup>45-47</sup>, with the central estimate of this factor between 2 and 3. The 2-minute average concentrations recorded in this study were multiplied by 0.4 based on these prior studies, and also based on comparisons of measurements made with three of the DustTraks from the apartment study with co-located gravimetric measurements when the units were deployed in a coincident study<sup>48</sup>. Table 1 provides summary statistics for the indoor and outdoor measurements for each apartment. Time-series of analyzed data from all apartments are presented in the SI. The adjusted indoor PM<sub>2.5</sub> concentrations reported by Norris et al. are in line with published PM<sub>2.5</sub> data from other studies of low-income homes in the U.S.<sup>49, 50</sup>, as discussed in the SI.

The data were analyzed to determine particle emission event characteristics including date, day of week, start and end time of emissions, identifier for linked events, estimated total removal rate including ventilation ( $\text{h}^{-1}$ ), estimated total mass emitted (mg) and emission rate (mg/min). The event characteristics were compiled into a database with other information including apartment and complex identifier, number of residents, number of bedrooms, and home volume. In addition to PM<sub>2.5</sub> concentrations, time resolved indoor and outdoor temperature, relative humidity (RH) and carbon dioxide (CO<sub>2</sub>) concentrations were measured in each of the 18 apartments and mean values of these parameters were calculated for each emission event.

## **Step 2. Calculate Infiltration Factor and Baseline Indoor Concentration of Outdoor Particles**

A baseline representing the indoor concentration of outdoor PM<sub>2.5</sub> was calculated with a variation of the method used by MacNeill et al.<sup>12</sup> to determine contributions of indoor and outdoor sources to fine PM concentrations in Canadian homes. The indoor-from-outdoor (IFO) baseline was calculated as the product of an infiltration factor ( $F_{inf}$ ) determined for each apartment and the time-varying outdoor concentration. The approach is described in detail and results, including sensitivity to the selected analysis parameters, are provided in the SI.

## **Step 3. Smooth Indoor Data and Identify Peaks Indicating Emission Events**

We used the R programming language MALDIquant package to smooth data and identify indoor PM<sub>2.5</sub> peaks<sup>51</sup>. The MALDIquant package was developed for analysis of chromatograph peaks, which presents the same core analytical challenge: to identify the beginning and end of a perturbation to the baseline signal in the presence of data noise. Real-time PM<sub>2.5</sub> concentrations, such as the 2-minute average data used in this study (Figure S1), contain data noise because of imperfect mixing and other factors.

An example application is shown in Figure 1. Figure 1a shows the 2-minute resolved PM<sub>2.5</sub> raw and smoothed data. Data were smoothed with the MALDIquant Savitzky Golay method, using a 3<sup>rd</sup> order polynomial fit and half-window size of 5 (11-point averaging) to dampen noise while preserving peak shape. Peak maxima – shown as vertical red lines in Figure 1b – were identified with the Detectpeak function using a half-window size of 5 and a signal-to-noise ratio of 2 or less, as required to ensure that peaks >10  $\mu\text{g}/\text{m}^3$  above the baseline were identified. Each identified peak was assumed to represent the end of the emission period and the beginning of post-emission decay. Figure 1b also shows the calculated baseline. Events where the peak PM<sub>2.5</sub> was <5  $\mu\text{g}/\text{m}^3$  above the IFO baseline were removed from the analysis because the small increase may not have resulted from indoor emissions.

## **Step 4. Identify Emission Event Start Times and Decay Period End Times**

An initial event start time was identified by looking back from the peak to identify the earliest set of three consecutive measurements with zero or positive change from the previous datum; the earliest was set as the start. The end of the decay was identified as the last three consecutive intervals with zero or negative change following the peak. The decay data were used to derive the loss rate of the event, as discussed below. The emission period is shaded in red in Figure 1b, and the decay period is shaded in gray.

In some cases, the initial event start and decay period end times were adjusted to more accurately define the period of sharp increase and subsequent decrease in concentrations. Adjustments were made only if the adjusted event start and/or decay period end times resulted in  $R^2 > 0.8$  from fitting of the emission rate and loss rate (Steps 7 and 8). Emission event start times were adjusted to the time just prior to the first point in the event when the indoor concentration was at least  $2 \mu\text{g}/\text{m}^3$  above the IFO baseline. Decay end times were reset as the last point that was at least  $2 \mu\text{g}/\text{m}^3$  above the baseline. The  $R^2 > 0.8$  condition resulted in start time adjustments of 310 peaks, of which the start time of 217 peaks was delayed to exclude concentrations  $< 2 \mu\text{g}/\text{m}^3$  above the IFO baseline when fitting the emission rate, and the start time of 93 peaks was moved forward to fully capture the entire emission period. Adjustments to decay period end time were made to 326 peaks, of which the decay period of 193 peaks was shortened to exclude concentrations  $< 2 \mu\text{g}/\text{m}^3$  above the IFO baseline when fitting the loss rate, and the decay period of 133 peaks were prolonged to fully capture the entire decay period (for example, see Figure 1c). If the concentration did not increase by at least  $5 \mu\text{g}/\text{m}^3$  from the event start to the peak, the event was removed from the database. This resulted in removal of 138 peaks identified by the Detectpeak function (29 of these were added back on visual review, as described in Step 6).

### **Step 5. Identify Linked Events**

Some peaks were close enough in succession that the end of the decay period of the first was the same point as the start of the next emission event; such peaks were considered “linked”. Linked events are identified because they may represent related emission sources. An example of two “linked” peaks is shown in Figure 1c. Peaks were de-linked if by the start of an event (i) the concentration dropped more than  $50 \mu\text{g}/\text{m}^3$  below the prior peak concentration or (ii) the concentration dropped by more than 50% of the peak concentration minus the IFO baseline.

### **Step 6. Visually Review and Add Peaks**

To facilitate visual review, we highlighted all indoor smoothed data that were  $> 10 \mu\text{g}/\text{m}^3$  above the baseline but not included in the emission or decay period of any identified event. There were 78 segments of data meeting these criteria. Those that followed a pattern of a relatively steep rise followed by decay towards the baseline were added to the event database; this included 29 that were identified by Detectpeak and removed because the peak was not  $> 5 \mu\text{g}/\text{m}^3$  above the starting concentration and another 17 small peaks that were missed entirely by Detectpeak. The remaining segments did not present as clear peaks or were too small to impact time-integrated PM in the home. Steps 4-5 were repeated for the added events.

In addition, we reviewed 10 peaks that had an emission duration lasting longer than 90 minutes for potential error made when adjusting the event start time. We restored the start time of 3 peaks to the



earliest of three consecutive measurements with zero or positive change. Visual review confirmed that the start time of the other 7 peaks were correctly identified so no correction was made.

In conducting the visual review, we also identified two cases of a saw-tooth pattern over a general decay that suggested removal through intermittent filtration. Peaks associated with the saw-tooth were removed.

### **Step 7. Determine Loss (Decay) Rates**

Indoor PM<sub>2.5</sub> loss processes include advection to outdoors through whole house and task ventilation, removal by filtration and deposition in mechanical air moving systems, and deposition and volatilization (sometimes associated with chemical transformations) in the indoor space<sup>52</sup>. Ventilation rates can change rapidly with operation of mechanical equipment – including exhaust fans, clothes dryers, and venting combustion appliances – or opening of windows and doors. Infiltration-driven air exchange generally changes over a time scale of hours with variations in outdoor temperatures and wind. The composite PM<sub>2.5</sub> loss rate from deposition and transformation processes in a given home can vary over time as it depends on aerosol characteristics including size distribution and size-dependent chemical composition, and environmental factors including air temperature, relative humidity, interior surface temperatures, air velocities, and mechanical filtration.

To calculate the emission rate for an event, we needed an estimate of the overall loss rate during the emission period. We determined the loss rate by fitting the smoothed PM<sub>2.5</sub> time series during either the 1st hour of the decay period or the entire decay period if it was shorter than 1 hour.

The following first-order mass balance equation was used to solve for the event specific, composite PM<sub>2.5</sub> pseudo-first order loss rate ( $L$ ):

$$\frac{dC_i}{dt} = -LC_i + PA C_{out} + \frac{E}{V} \quad (1)$$

In this equation,  $C_{in}$  is the indoor concentration,  $C_{out}$  is the outdoor concentration,  $P$  is the penetration factor (the net fraction of outdoor PM<sub>2.5</sub> that is not removed as outdoor air enters the residence through all pathways),  $A$  is the air exchange rate,  $V$  is the mixing volume, and  $E$  is the PM<sub>2.5</sub> emission rate indoors.

During the decay period, with  $E = 0$ , the general solution is provided in Eq. 2,

$$C_i(t) - C_{i_o} = (C_i(t_d) - C_{i_o}) * \exp(-L(t - t_d)) \quad (2)$$

where  $C_{in}(t_d)$  is the concentration at the beginning of the decay period.  $C_{in_o}$  is the indoor PM<sub>2.5</sub> concentration from the entry of outdoor PM<sub>2.5</sub>, taken as the average IFO baseline during the emission event. The loss rate  $L$  is determined by the slope of a linear fit to the data, as follows:

$$\ln \left( \frac{C_i(t) - C_{i_0}}{C_i(t_d) - C_{i_0}} \right) = -L(t - t_0) \quad (3)$$

### Step 8. Calculate PM<sub>2.5</sub> Event Emission Rates

Once the loss rate was calculated for each event, the emission rate was calculated using Eq. 4.

$$d \frac{(C_i(t) - C_{i_0})}{dt} = \frac{E}{V} - L C_i \quad (4)$$

The emission rate,  $E$ , is determined by fitting the measured data to a linear model corresponding to the general solution (Eq. 5), with the values of  $L$  and  $C_{in,0}$  determined as described above and taking  $C_{in}(t_0)$  as the concentration at the beginning of the emission event.

$$\overbrace{(C_i(t) - C_{i_0}) - (C_i(t_0) - C_{i_0}) \exp(-L(t - t_0))}^y = \overbrace{E \left[ \frac{1}{LV} (1 - \exp(-L(t - t_0))) \right]}^m \quad (5)$$

The slope ( $m$ ) determined from the linear model fit is the event emission rate,  $E$ . For example, Figure 1d shows the PM<sub>2.5</sub> concentrations modeled using the calculated emission and decay rates.

Equations 4-5 assume that the loss rate during the decay applies during the emission period. This induces an error in the calculated emission rate when the loss rate changes. Coagulation and gas-particle partitioning processes are of particular relevance to this concern as partitioning changes total aerosol mass and both processes affect aerosol size distribution, which in turn affects the aggregate deposition rate.

### Compare Simulated to Measured PM<sub>2.5</sub> Concentrations During Events

To evaluate the degree to which the ideal model produced results consistent with the measurements, we calculated concentrations in each apartment using the values of  $E$  and  $L$  determined for each event and compared this simulated time series to the measured data. The simulated and measured time series are shown in Figure S1 of the SI.

### Compare to Method Used by Wallace et al. (2006)

As a point of comparison to prior work, the apartment data were also analyzed using the method described in an analysis of data from 37 homes in North Carolina<sup>14</sup>. Wallace started by calculating a 4-minute running average then set the event start as an increase of 7  $\mu\text{g}/\text{m}^3$  from one data point to the next.

When the running average fell to  $<10 \mu\text{g}/\text{m}^3$  above baseline concentration (determined as the indoor concentration immediately preceding the event start), the peak continued until the first minute when there was an increase in the concentration.

## 3 Results and Discussion

### 3.1 Identified event characteristics

A total of 836 emission events were identified and quantified from 224 days of monitoring data. Summary statistics for estimated  $\text{PM}_{2.5}$  mass emitted, event duration, emission rate, and pseudo-first order decay rate for all characterized events are presented in Table 2. Except for event duration, the parameters were approximately lognormally distributed (see Figure S5). Even with the thresholds to eliminate very small events, most quantified  $\text{PM}_{2.5}$  emission events were small in mass emitted (median = 12 mg) and relatively brief in duration (median = 16 minutes).

The  $\text{PM}_{2.5}$  emission rates and emitted masses determined for events in this study fall within the very large range of residential, activity-related  $\text{PM}_{2.5}$  emissions reported in prior studies<sup>16, 19, 21, 24, 42, 53</sup>, as discussed in the SI. This literature indicates that certain cooking activities emit more  $\text{PM}_{2.5}$  per event and have higher emission rates than most of the non-cooking related particle-producing activities that have been studied, with the exception of smoking. The statistics for events analyzed in the current study are consistent with a mix of cooking and non-cooking activities.

The event statistics characterized from our analysis included all peaks that were identified, regardless if they were “linked” or not. For most apartments (2-1, 2-4, and 3-5 are the exceptions, see Figure S1), 80% or more of the identified emission events are isolated peaks. “Linked” peaks may represent multiple emission events generated by one larger activity, e.g. several distinct cooking activities as part of preparing one meal. Of the 836 peaks identified, 325 of them are characterized as linked peaks. All together, they formed 117 linked events. Linked events are important because with longer overall emission duration and more mass emitted in succession, linked events tend to result in higher peak indoor  $\text{PM}_{2.5}$  concentrations compared to events composed of isolated peaks.

The distribution of first order decay rates determined for events in this study were similar to loss rates determined in a 13-home study in Australia<sup>54</sup>, but higher than those reported for large studies in North Carolina, USA<sup>42</sup> and Edmonton, Canada<sup>55</sup>; details are presented in the SI. Our use of an overall first-order decay model to approximate the combined effect of all loss mechanism is supported by the high  $R^2$  that resulted from the fitting. About 95% of the identified emission events had  $R^2 > 0.8$ . We considered that very high fitted loss rates could indicate that the monitor was close to the source and the rapid decay would then reflect mixing and dilution to the complete home volume. When this occurs, there should be

an initial period of sharp decay followed by a period of much slower first order decay. We observed an inverse relationship between loss rate and duration of the decay period from which the loss rates were fitted. Among the 836 peaks identified, 11 peaks had a loss rate  $>10/h$ ; all were determined from a very brief decay period (8 to 16 minutes, median = 10 minutes). There were 52 peaks that had a loss rate between  $5/h$  and  $10/h$ ; their decay period also tended to be shorter (6 to 48 minutes, median = 15 minutes) than peaks that had a loss rate  $<5/h$  ( $N=774$ ), where the decay period ranged between 2 and 680 minutes (median = 48 minutes).

Table 3 shows the calculated contributions of indoor events to overall  $PM_{2.5}$  concentrations. The time-integrated indoor  $PM_{2.5}$  contributed by the characterized emission events varied from 15 to 86% with an average of 56% across apartments. At the low end of previously reported values, Allen et al.<sup>10</sup> estimated that non-ambient (i.e., indoor) sources on average accounted for only 21% of indoor  $PM_{2.5}$  concentrations in 44 homes in Seattle, Washington; however, the authors indicated that the elderly residents who populated the majority of homes in that study were less active than residents of other studies, citing Liu et al.<sup>56</sup>. Meng et al.<sup>57</sup> reported non-ambient contributions of 33%, 30%, and 59%, respectively, to indoor  $PM_{2.5}$  measured in a total of 212 non-smoking homes in California, New Jersey, and Texas. Wallace et al.<sup>14</sup> reported that ambient sources contributed about half of the total  $PM_{2.5}$  measured in the homes of 37 health-compromised subjects in North Carolina. And Habre et al.<sup>13</sup> found that 72% of the  $PM_{2.5}$  measured in the homes of 37 asthmatic children in New York City was attributable to indoor sources.

Table 3 also shows that using the idealized parameters determined from the fitting algorithm to simulate events yields integrated concentrations similar to the actual measured time series. Table 3 presents the means of the event mean and highest 10-minute  $PM_{2.5}$  indoor concentrations during the characterized emission events for each apartment. Across all events identified,  $PM_{2.5}$  concentrations calculated using the event parameters produced event means that were 3.0% higher on average ( $SD = 7.5\%$ ), and highest 10-minute concentrations during events that were 6.1% lower on average ( $SD = 7.5\%$ ), compared with corresponding measurements during events. With just one exception, the means of the ideally modeled peaks agreed to within 15% of the measured values for each apartment. Figures S3 and S4 in the SI present modeled to measured comparison for each apartment. These comparisons show that the ideal model is able to quantitatively reproduce the measurements.

### **3.2 Diurnal variability in event characteristics**

To assess the impact of time of day on event occurrence and emitted mass, each hour of the day was classified as either with or without indoor emissions. Figure 2 shows the fraction of all hours of available data that are classified as either having or not having an indoor emission event. Indoor emissions tended to occur most frequently in the afternoon and evening between 1400 and 2200 hours. The hours with the

highest occurrence were 1700-2100, indicating the importance of dinner cooking. Hours between 0200 and 0600 had the lowest occurrence of indoor emissions, as expected during the time when most people are asleep. The overall pattern was similar for weekdays and weekend days. There was a large variation in the event occurrence across apartments, as described in Figure S6, likely reflective of differences in occupancy patterns and activity levels. This could not be confirmed because there are no occupancy or activity data.

Figure 3 shows the mean emitted mass by hour of the day across all apartments. There are large differences in the time-of-day pattern and magnitude of emitted mass across the apartments. Two of the apartments had hours in which >50 mg of PM<sub>2.5</sub> were emitted on average, while other apartments had no hours with >5 mg of PM<sub>2.5</sub> emitted. Some apartments had emissions only during evening hours, while others had most emissions occurring in the middle of the day. These differences are consistent with variations in occupancy schedule and activities across homes.

Figure 4 shows the range of event duration and loss rate for identified emission events, as a function of time of day. Loss rates are plotted during the hour when the emission event had just ended (i.e., at the time of the peak). The majority of the emission events (77%) lasted for 10 to 30 minutes. Emission events lasting longer than 30 minutes tend to occur more frequently during hours that correspond to meal preparation times (0070 and 0080 for breakfast, 0012 and 0013 for lunch, and 0017 and 0018 for dinner). Hours with high loss rate (>5/h) tend to occur during early morning (0060 and 0070), early afternoon (0014 and 0015), and during evening hours (0017 to 0019).

### **3.3 Events identified using method of Wallace et al. (2006)**

Applying the analysis algorithm described by Wallace et al.<sup>14</sup> identified 312 emission events that accounted for an average of 36% of the time-integrated PM<sub>2.5</sub> measured in the 18 homes, i.e. many fewer events and a much smaller fraction than identified by the approach of this study. Additional details are provided in the SI. In comparison, our method identifies more peaks, especially in apartments with many linked events (e.g., 2-1, 2-4, and 3-5) consisting of multiple distinct peaks that the method by Wallace et al. would combine as one continuing event. Our method also captures higher contributions of emission events to indoor PM<sub>2.5</sub> in apartments (Complex 1) with smaller peaks that the method by Wallace et al. missed.

### **3.4 Relationships between event characteristics**

We used the non-parametric Spearman correlation to identify statistical associations for pairs of event characteristics and between event and household characteristics, with results presented in Table 4. We did not use the more common Pearson's correlation because the relationships are not necessarily linear and

because the Pearson's correlation is very sensitive to outliers. The Spearman correlation statistic indicates the degree to which a monotonic function can describe the relationship between two parameters; positive values indicate two parameters increasing together, negative values indicate one parameter decreasing as the other increases. Table 4 shows six pairs of parameters with correlation coefficients above 0.2 and statistically significant at  $p < 0.05$ .

Emitted mass was positively correlated with event duration and loss rate. The first relationship is intuitive and rational: longer events were associated with more  $PM_{2.5}$  being emitted. The association between emitted mass and loss rate results in part from the mass balance equation used to calculate the emission rate: a higher decay rate, which was determined first, directly translates to a higher emission rate. Emitted mass was positively correlated with house volume and the difference between the indoor and outdoor carbon dioxide, and also positively, if weakly correlated with the number of residents and number of bedrooms. These parameters are all related to actual or expected occupancy, suggesting that more people in the home translates to larger emission events. As with loss rate, the relationship to house volume – which had the highest correlation coefficient of the four – could be partly an artifact of the mass balance equation used to calculate mass. There were weak correlations between emitted mass and indoor RH and outdoor temperature. These associations could reflect differences across complexes, rather than just environment. Complexes 2 and 3 were measured in winter (mean outdoor temperature of 9 °C during emission events) and had much higher emissions (see Figure 3) than apartments in Complex 1, which were measured in the summer (mean outdoor temperature of 24 °C during events). Differences across complexes also likely explain the weak correlations between indoor temperature and event duration. More data would be needed to assess seasonal effects independent of potential trends by building complex.

Loss rate was negatively correlated with event duration. This relationship is expected for short emission events that also tend to have a shorter decay period, with mixing throughout the home producing a sharp initial decay rate. The loss rate was positively correlated with indoor and outdoor temperature and negatively correlated with indoor and outdoor RH. But as with the relationships of these parameters with emitted mass, these associations could reflect differences across complexes, rather than just environment. A small, negative correlation with  $CO_2$  indoor-outdoor difference was seen. These environmental conditions correspond to warmer weather and may be associated with more window opening, which should produce higher loss rates than closed windows. We hypothesize that imperfect mixing more commonly impacted events in apartments with more bedrooms, as there would be a higher likelihood that some emission sources were not in the central zone where  $PM_{2.5}$  was measured.

### 3.5 Limitations

There are important limitations to both the general method presented for indoor PM<sub>2.5</sub> emission event characterization and the specific application described here. The first is that the data analyzed in this study – and most of the available time-resolved data on PM<sub>2.5</sub> concentrations in homes – were obtained using light-scattering monitors that estimate, but do not directly measure PM<sub>2.5</sub>. Since these devices are commonly calibrated with standardized aerosols (e.g., Arizona Road Dust), which may have very different scattering properties compared to many indoor-generated aerosols, there is a need to adjust the measured data. Adjustments based on side-by-side measurements in residential environments, or in controlled experiments with residential sources, should provide mass emission rates much closer to those determined with a gravimetric reference or equivalent method. However, aerosols generated from gas to particle conversion processes – including candles, volatilization of organics from hot surfaces<sup>58</sup>, and many cooking-related emissions – may have substantial mass below the minimal detectable particle size for a light scattering device, resulting in a biased measurement of such sources. For example, a study with the TSI Sidepak Model AM510 ([www.tsi.com](http://www.tsi.com)) reported ratios of gravimetric to scattering-based measurements for varied indoor sources<sup>59</sup>: 0.32 for cigarettes, 0.35 for stick incense, 0.44–0.47 for fireplace emissions, 0.41–0.70 for various cooking activities.

The data analyzed for this study did not include occupancy data or activity logs; as a result, the identified emission events could not be attributed to specific activities or occupancy at the time of the event. The results would be more valuable if the general ventilation conditions, e.g. windows open or closed, were included in the event database. Knowledge about the timing of any large increases in ventilation that correspond with an increase in indoor concentrations toward outdoor levels would distinguish those situations from indoor generation events. Ventilation targeted to source reduction, e.g., using a range hood, should produce lower estimates of emission rates and emitted mass than would occur without venting. This should not be regarded as an error since venting can actually reduce the amount of material introduced into the mixing volume of the home. Any use of event data from the current study to assess potential benefits of mitigation options should consider this limitation.

The method underestimates the total number and frequency of emission events because small magnitude deviations from baseline PM<sub>2.5</sub> time-series trends are explicitly excluded. Two or more activities occurring very close in time may appear as a single event.

The assumption of instantaneous and uniform mixing produces biases when the PM source is close to the detector or there is directional airflow in the building that leads to removal of the source before complete mixing occurs. If the relevant mixing volume during an event is less than the total residence volume and both the source and measurement device are in that mixing volume, both the emission rate and emitted

mass will be biased high by the ratio of the total volume to the initial mixing volume. In such cases, mixing will present as a higher decay rate than would be appropriate to the entire residence. The ideal model also assumes constant emission and decay rates and does not examine the impact of variations in these parameters during an event. For cases in which concentrations begin to decrease because the source emission rate declined (with emissions continuing at a lower level), both the decay rate and event duration would be underestimated. As long as the integral of the modeled concentration reasonably approximates the integral of the measured concentration over the course of the event, using the determined event characteristics in modeling should provide an accurate estimate of exposure.

The analysis algorithm used in the present study specifies all parameters and thresholds needed to identify and characterize events. Of the prior studies that analyzed emission event characteristics, only Wallace et al.<sup>14</sup> reported the thresholds used to identify event start and end times. Olson et al.<sup>42</sup> reported criteria for event identification but not precise start and end times. Other studies that reported on particle emission events<sup>19, 20</sup> did not specify the criteria used. It is uncertain if the thresholds used to identify events in the current study will work for other data sets.

### **3.6 Predicting emission event characteristics for modeling indoor exposure**

Identified event schedules and characteristics can be used to predict indoor PM<sub>2.5</sub> concentration patterns in residential modeling. The established diurnal frequency of events can be used as the probability that an event will occur during each hour of the day. Once it has been established that an event occurs, the characteristics of the event need to be simulated as well. For each event, the emitted mass, duration, and loss rate can be selected from fitted lognormal distributions for each parameter. Linked events can be modeled as sequences such that the simulation will capture the evaluated indoor PM<sub>2.5</sub> concentrations resulting from indoor emissions occurring in succession.

## **4 Conclusions**

This study demonstrated an approach to identify and characterize residential PM<sub>2.5</sub> emission events that utilizes an automated analysis tool developed for chromatographic peak analysis, supplemented by visual review to confirm that all apparent events are identified. The approach was applied to characterize all emission events occurring over 5–14 days of monitoring in 18 California apartments with low-income residents. Average event frequencies across homes showed little weekday-weekend differentiation. Emitted mass was positively correlated with event duration and loss rate. Emitted mass was also positively correlated with number of residents, number of bedrooms, house volume, and the difference between indoor and outdoor CO<sub>2</sub>. Lognormal distributions of emitted mass, event duration, and loss rate



can be used to model indoor  $PM_{2.5}$  exposures stochastically with probabilities derived from the diurnal frequency of events.

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## Figures and Tables

**Table 1. Characteristics of 18 California apartments for which PM<sub>2.5</sub> emission events were characterized.<sup>a</sup>**

Complex-Apartment	Stove Type	Residents	Bedrooms	Floor Area (m <sup>2</sup> )	Days of PM <sub>2.5</sub> Data	Mean PM <sub>2.5</sub> (µg/m <sup>3</sup> )	
						Indoor	Outdoor
1-1 <sup>b</sup>	Gas	4	4	92	13.6	5.4	2.2
1-2 <sup>b</sup>	Gas	3	3	85	13.8	1.6	2.2
1-3 <sup>b</sup>	Gas	6	4	92	13.8	4.1	2.2
1-4 <sup>b</sup>	Gas	4	3	85	13.8	1.8	2.2
1-5 <sup>b</sup>	Gas	5	4	92	13.8	1.9	2.2
1-6 <sup>b</sup>	Gas	5	3	85	7.0	2.5	2.1
2-1	Gas	1	1	67	13.0	16.4	10.4
2-2	Gas	2	2	76	12.7	5.6	10.4
2-3 <sup>b</sup>	Gas	4	3	125	10.7	42.5	10.4
2-4 <sup>b</sup>	Gas	3	3	125	13.8	29.4	10.2
2-5 <sup>b</sup>	Gas	3	3	125	12.9	14.0	10.5
2-6 <sup>b</sup>	Gas	7	4	139	5.2	39.4	11.1

3-1	Elec.	2	2	80	13.0	8.2	20.9
3-2	Elec.	1	2	80	13.9	17.7	22.1
3-3	Elec.	5	2	80	13.9	13.7	22.1
3-4	Elec.	3	2	80	12.8	52.5	20.8
3-5	Elec.	4	3	98	13.8	64.1	22.0
3-6	Elec.	4	3	98	12.8	13.6	20.9

<sup>a</sup> Complexes were located in Sacramento (1), Richmond (2), and Fresno (3). Statistics of PM<sub>2.5</sub> (days of monitoring, mean concentrations) refer to the pre-retrofit period only.

<sup>b</sup> Indicates two stories; all other apartments are single story.

**Table 2. Summary statistics for the 836 fine particle emission events identified from time-resolved data collected in 18 low-income apartments in California.**

	Mean	Median	Geometric Mean	Geometric Std. Dev.	5 <sup>th</sup> to 95 <sup>th</sup> Percentile Range
Emitted Mass (mg)	30	12	13	3.7	1.4 to 154
Event Duration (minutes)	23	16	19	1.7	8 to 66
Emission Rate (mg/h)	103	37	40	3.9	3 to 582
Loss Rate (1/h)	2.0	1.3	1.3	2.5	0.2 to 7.5



**Table 3. Summary statistics for measured and modeled PM<sub>2.5</sub> concentrations during identified emission events in each apartment.**

Complex -Apt	Number of Events	Contribution of Events to Indoor PM <sub>2.5</sub>	Event Averaged PM <sub>2.5</sub> During Emission Events			Highest 10-Minute Average PM <sub>2.5</sub> During Emission Events		
			Measured Mean ( $\mu\text{g}/\text{m}^3$ )	Modeled Mean ( $\mu\text{g}/\text{m}^3$ )	% Diff.	Measured Mean ( $\mu\text{g}/\text{m}^3$ )	Modeled Mean ( $\mu\text{g}/\text{m}^3$ )	% Diff.
1-1	38	69%	17.1	16.7	-3%	43.3	36.0	-17%
1-2	5	17%	10.9	10.9	-0.1%	19.9	18.5	-7%
1-3	20	60%	14.6	13.8	-5%	52.1	45.2	-13%
1-4	11	35%	11.4	13.1	15%	39.2	39.9	2%
1-5	14	21%	9.7	10.1	5%	21.0	20.2	-4%
1-6	11	36%	7.8	9.2	18%	19.7	19.9	1%
2-1	47	69%	44.2	44.3	0.3%	70.1	67.9	-3%
2-2	20	40%	31.0	31.5	1%	63.5	61.9	-3%
2-3	102	86%	76.7	78.1	2%	168.2	154.0	-8%
2-4	99	77%	55.4	56.0	1%	98.1	94.5	-4%
2-5	42	67%	29.4	31.1	6%	96.7	95.1	-2%

2-6	34	79%	80.0	90.5	13%	196.6	196.0	-0.3%
3-1	6	15%	18.7	15.9	-15%	45.9	32.4	-29%
3-2	36	75%	74.1	78.3	6%	145.3	136.3	-6%
3-3	20	56%	47.4	48.5	2%	115.5	114.4	-1%
3-4	132	73%	112.1	114.2	2%	188.9	177.3	-6%
3-5	159	82%	94.8	97.6	3%	134.0	132.0	-1%
3-6	40	43%	33.6	34.3	2%	71.8	65.5	-9%

**Table 4. Spearman correlation coefficients<sup>a</sup> between characteristics determined for indoor emission events and environmental conditions measured during the events.**

	Emitted Mass	Event Duration	Loss Rate
Event Duration	<b>0.17 (&lt;0.005)</b>		
Loss Rate	<b>0.29* (&lt;0.005)</b>	<b>-0.30* (&lt;0.005)</b>	
Number of Residents	<b>0.10 (&lt;0.005)</b>	-0.03 (0.40)	0.05 (0.13)
Number of Bedrooms	<b>0.08 (0.01)</b>	-0.04 (0.24)	<b>0.11 (&lt;0.005)</b>
Home Volume	<b>0.29* (&lt;0.005)</b>	-0.02 (0.55)	0.03 (0.32)
CO <sub>2</sub> Indoor-Outdoor <sup>b</sup>	<b>0.27* (&lt;0.005)</b>	-0.06 (0.16)	<b>-0.1 (0.02)</b>

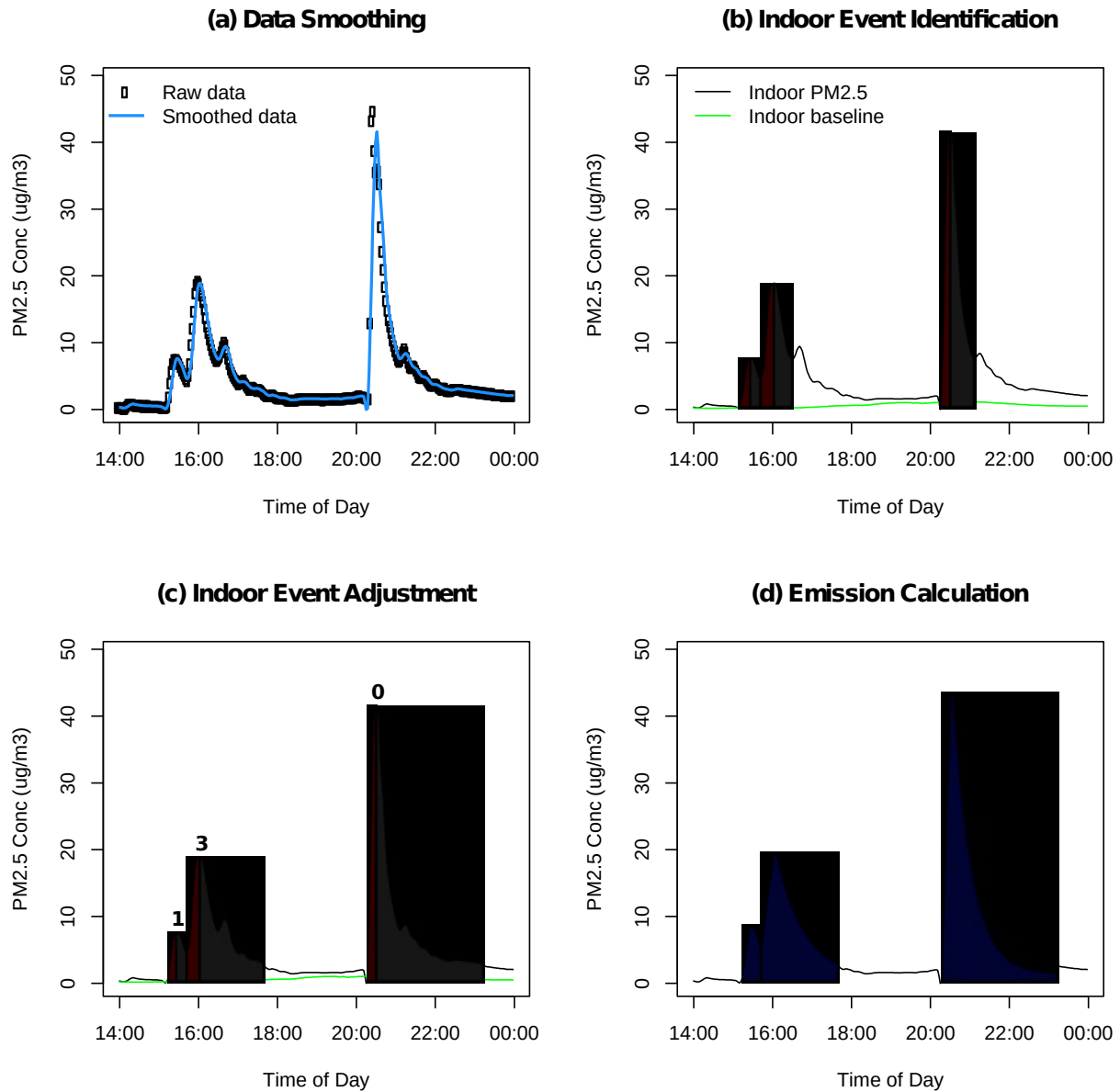
RH Indoor	<b>0.08 (0.02)</b>	0.03 (0.40)	<b>-0.25* (&lt;0.005)</b>
RH Outdoor	-0.06 (0.10)	0.01 (0.86)	<b>-0.15 (&lt;0.005)</b>
Temperature Indoor	-0.05 (0.15)	<b>-0.10 (0.009)</b>	<b>0.20* (&lt;0.005)</b>
Temperature Outdoor	<b>-0.14 (&lt;0.005)</b>	-0.05 (0.13)	<b>0.18 (&lt;0.005)</b>

<sup>a</sup> Correlation coefficients with significance (shown in parentheses) of  $p < 0.05$  levels are in bold.

<sup>b</sup> During the event.

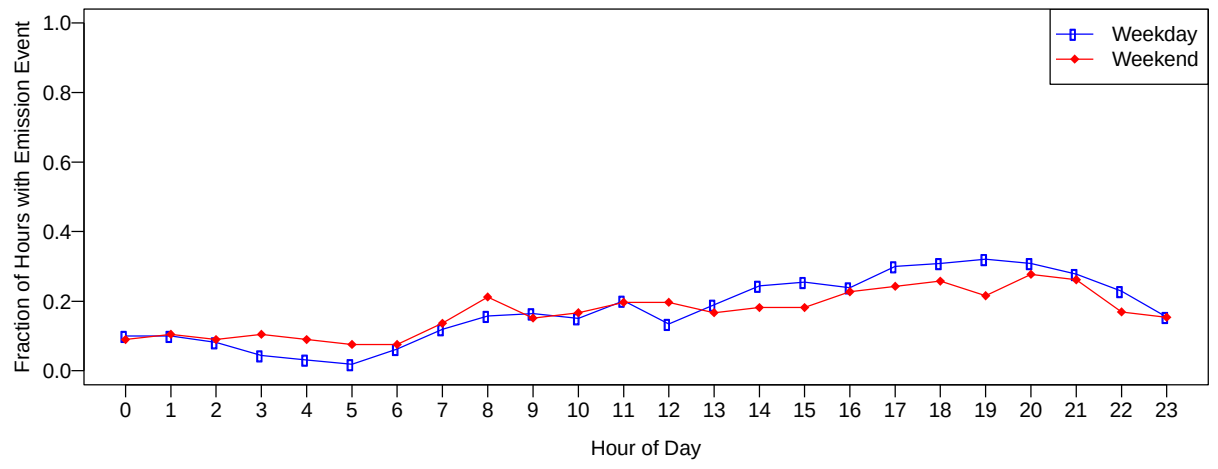
\*Asterisk indicates pairs with a correlation coefficient above 0.2.

**Figure 1. Steps in the identification and characterization of indoor emission events using R programming language MALDIquant package.** Adjustments of start and end time, and whether indoor events are considered “linked” (peaks labeled 1 and 3 in (c) are “linked”, peak 0 is not), were determined using the indoor baseline. Panel (d) shows the profile associated with the constant emissions and first order decay models used to determine event characteristics.



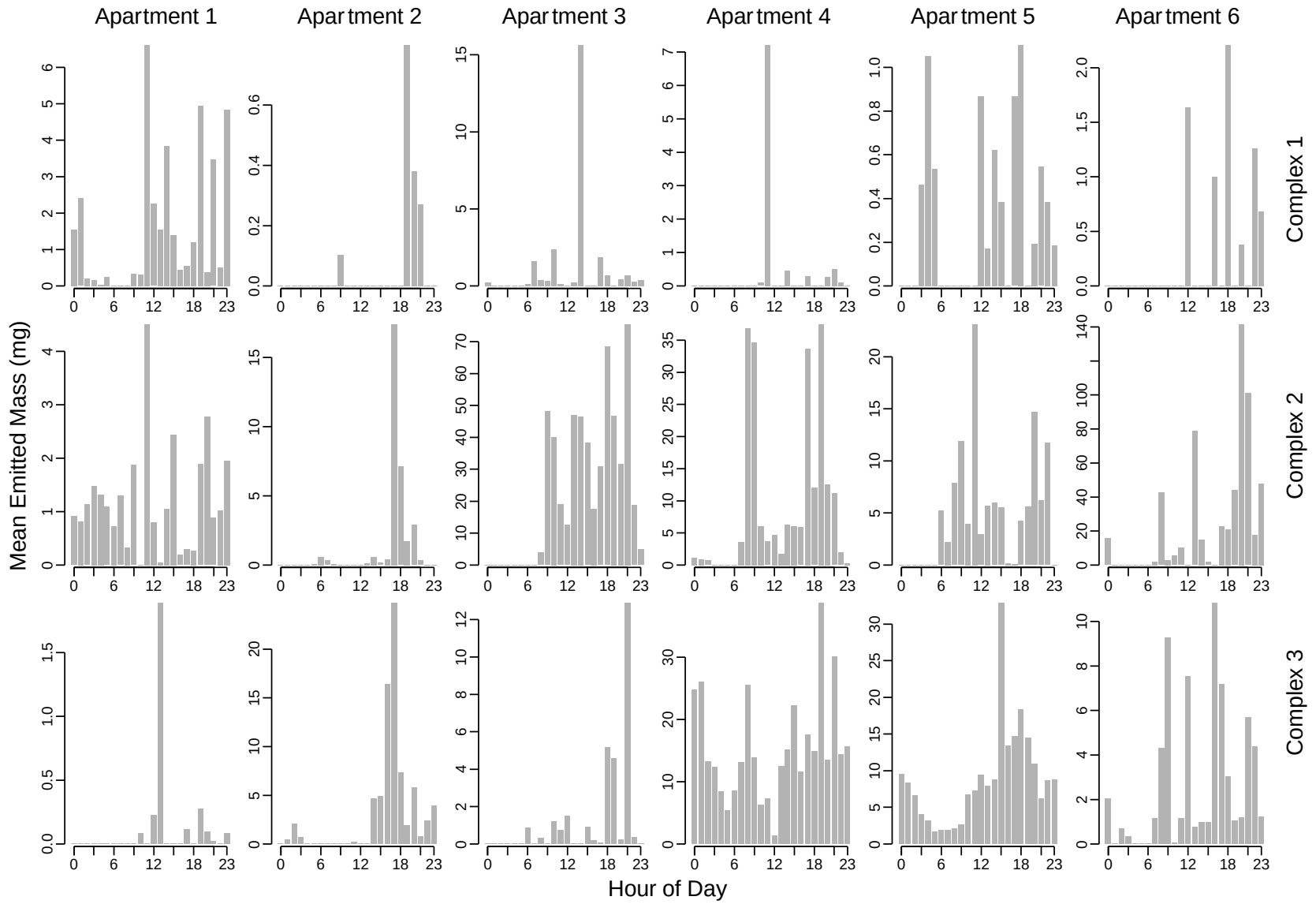


**Figure 2. Fraction of hours with any identified emission events across 228 total days of monitoring in 18 California low-income apartments.**

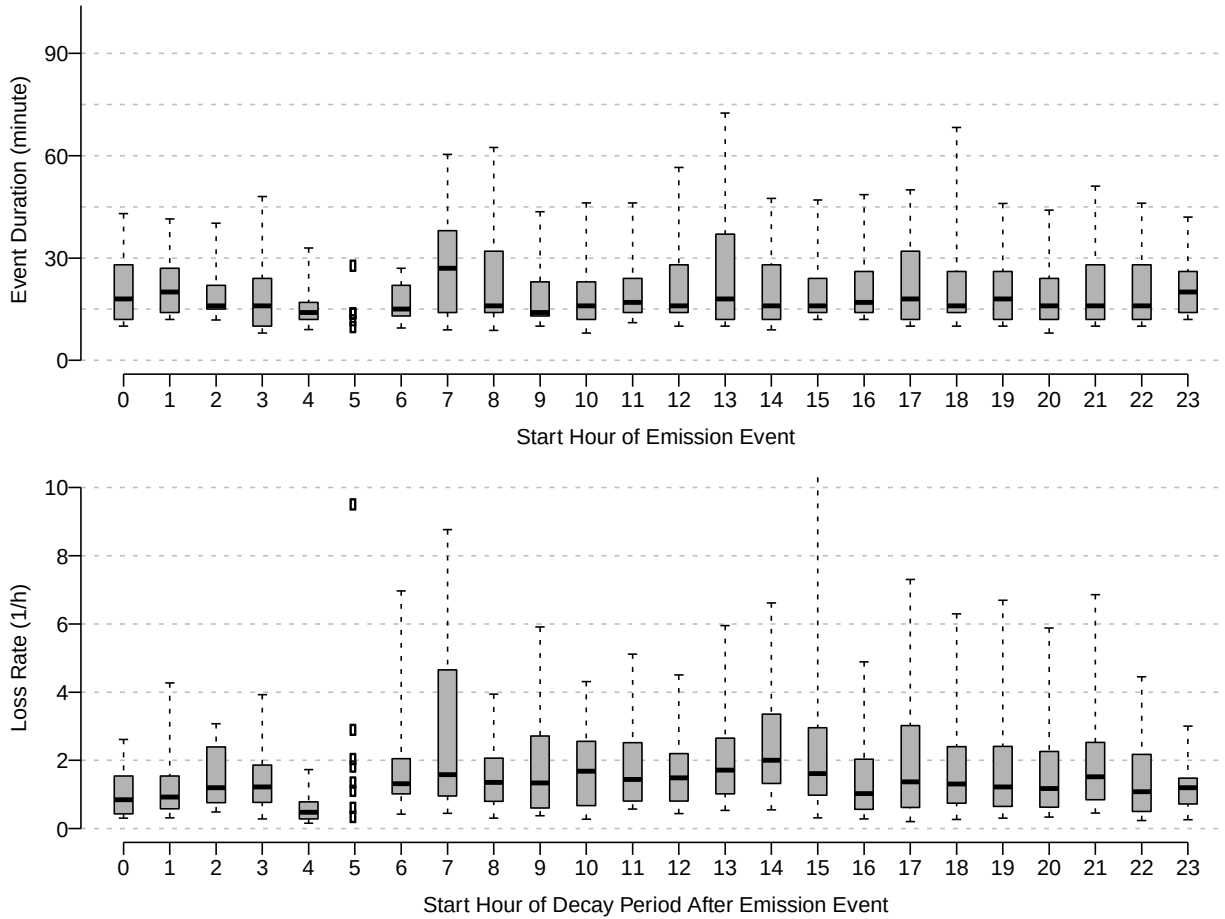


for each hour of the day for the 18 California apartments.

Figure 3. Mean emitted mass of  $PM_{2.5}$



**Figure 4. Duration and calculated loss rate for identified indoor emission events by hour of day.** The box shows median and the interquartile range (25<sup>th</sup> to 75<sup>th</sup> percentile), and the whiskers extend to 5<sup>th</sup> and 95<sup>th</sup> percentiles. Circles present all data during hours with fewer than 10 events. For some events that started during Hour 7, the decays did not start until Hour 8.



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