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Healthy Zero Energy Buildings (HZEB) Program – Cross Sectional Study of Contaminant Levels, Source, Strengths, and Ventilation Rates in Retail Stores.

Wanyu R. Chan, Meera Sidheswaran, Sebastian Cohn, Douglas P. Sullivan, and William Fisk

Environmental Energy Technologies Division

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Final Report

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1 Abstract

This field study measured ventilation rates and indoor air quality parameters in 21 visits to retail stores in California. The data was collected to guide the development of new, science-based commercial building ventilation rate standards that balance the dual objectives of increasing energy efficiency and maintaining acceptable indoor air quality. Data collection occurred between September 2011 and March 2013. Three types of stores participated in this study: grocery stores, furniture/hardware stores, and apparel stores. Ventilation rates and indoor air contaminant concentrations were measured on a weekday, typically between 9 am and 6 pm. Ventilation rates measured using a tracer gas decay method exceeded the minimum requirement of California's Title 24 Standard in all but one store. Even though there was adequate ventilation according to Title 24, concentrations of formaldehyde, acetaldehyde, and acrolein exceeded the most stringent chronic health guidelines.

Other indoor air contaminants measured included carbon dioxide (CO_2), carbon monoxide (CO), ozone (O_3), and particulate matter (PM). Concentrations of CO_2 were kept low by adequate ventilation, and were assumed low also because the sampling occurred on a weekday when retail stores were less busy. CO concentrations were also low. The indoor-outdoor ratios of O_3 showed that the first-order loss rate may vary by store trade types and also by ventilation mode (mechanical versus natural). Analysis of fine and ultrafine PM measurements showed that a substantial portion of the particle mass in grocery stores with cooking-related emissions was in particles less than 0.3 μm. Stores without cooking as an indoor source had PM size distributions that were more similar indoors and outdoors.

The whole-building emission rates of volatile organic compounds (VOCs) and PM were estimated from the measured ventilation rates and indoor and outdoor contaminant concentrations. Mass balance models were then used to determine the ventilation rates, filtration strategies, or source reductions needed to maintain indoor contaminant concentrations below reference levels. Several scenarios of potential concern were considered: (i) formaldehyde levels in furniture/hardware stores, (ii) contaminants associated with cooking (e.g., PM, acrolein, and acetaldehyde) in grocery stores, and (iii) outdoor contaminants (e.g., PM and O_3) impacting stores that use natural ventilation. Estimated formaldehyde emission rates suggest that retail stores would need to ventilate at levels far exceeding the current Title 24 requirement to lower indoor concentrations below California's stringent formaldehyde reference level. Given the high costs of providing ventilation but only modest chronic health benefit is expected, effective source control is an attractive alternative, as demonstrated by some retail stores in this study. Predictions showed that grocery stores need MERV 13 air filters, instead of MERV 8 filters that are more commonly used, to maintain indoor PM at levels that meet the chronic health standards for PM. Exposure to acrolein is a potential health concern in grocery stores, and should be addressed by increasing the use of kitchen range hoods or improving their contaminant removal efficiency. In stores that rely on natural ventilation, indoor PM can be a health concern if the stores are located in areas with high outdoor PM. This concern may be addressed by switching to mechanical ventilation when the outdoor air quality is poor, while continuing natural ventilation when outdoor air quality is good.

2 Introduction

The Healthy Zero Energy Buildings (HZEB) research program aims to generate information needed to develop new science-based commercial building ventilation rate (VR) standards that balance the dual objectives of increasing energy efficiency and maintaining acceptable indoor air quality. This report describes the HZEB field study of retail stores. The primary purpose of this study is to estimate the whole-building source strengths of contaminant of concerns (COCs). This information is needed to determine the VRs necessary to maintain indoor concentrations of COCs below applicable health guidelines.

The California Commercial End-Use Survey (Itron, 2006) found that retail buildings account for about 15% of the commercial building energy use. Retail buildings consume the third largest share of total electricity consumption, following large offices (17%) and miscellaneous (16%). The survey estimated that about 14% of the total commercial floor stock in California belongs to retail buildings. Space cooling and ventilation account for about one-third of the electricity usage, which is the second predominant electricity end use in retail buildings, after interior lighting (43%). Retail also employs a large number of workers and the stores are frequently visited by the general public. The 2007 Economic Census (Census, 2009) shows that in California, the "retail trade" employs 1.7 million workers. Over 40% of the workers are employed by the store types that are considered in this study: grocery, furniture, hardware, and apparel stores. The National Human Activity Pattern Survey (NHAPS) found that of the time people spent indoors outside of their home, 13% is spent in shopping mall and grocery stores (Klepeis, 2001). It is the third most frequented place after offices (23%) and schools (22%).

California's building energy efficiency standards (Title 24) specifies retail stores to be ventilated at the larger of 7 L/s per person or 1 L/s-m^2, with the per floor area value often used for design purposes. To the best of our knowledge, these minimum ventilation rates (VRs) are based on professional judgment, such as from the American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE, 2010). It remains a question whether the minimum VRs are adequate or excessive for controlling contaminant levels. Studies have found the types of contaminants and their concentrations to vary considerably among different retail stores (Loh, 2006, Eklund, 2008, Wu, 2011). Consequently, the required VRs may also vary by store types. For example, Loh et al. (2006) found that furniture stores tend to have higher formaldehyde concentrations than other retail types. Wu et al. (2011) also found statistically significant differences by building types for formaldehyde and a number of other volatile organic compounds (VOCs), such as ethylbenzene, m/p-xylene, o-xylene, chloroform, tetrachloroethylene, and naphthalene.

The goal of this study is to identify contaminants in retail stores that may be controlled via ventilation, and to determine the minimum VRs that would satisfy the occupant health and odor criteria. Nineteen stores were sampled, of which two stores were sampled twice, thus making the total count to twenty-one store visits. To determine if contaminant source strengths differ by store types, three types of retail stores are included in this study: grocery, furniture/hardware, and apparel. The current minimum VR requirement of California Title 24 (CEC, 2008), as described above, applies to all these store types. A key focus of this work is on VOCs because there are numerous indoor sources, such as from building materials and merchandise. Also, indoor air

concentrations of many VOCs are highly affected by VRs, while concentrations of particles are less affected (Parthasarathy, 2013). Findings of this work may inform future VR standards in Title 24. The other indoor contaminants that were also measured in this study include carbon dioxide, carbon monoxide, ozone, fine and ultrafine particles.

3 Method

Nineteen stores were recruited from the various parts of California, including ten from the Bay Area (north/central coastal), three near Sacramento, one in Fresno, and five in the Los Angeles area (south coastal). Two stores were sampled twice. Field sampling started in September 2011 and ended in March 2013. Samples were collected for two consecutive days in the first three stores that participated in this study. Remaining stores were sampled for one day only such that given limited resources, more stores could be included in this field study. Stores volunteered to participate in this study. Stores were not selected based on building characteristics criteria, so they formed a convenience sample.

Besides collecting contaminant samples, basic information about the stores was also gathered. The number of occupants, including store workers and customers, were counted a few times during the day of sampling. Information about the heating, ventilation, and air conditioning (HVAC) system was noted, such as equipment model, operational mode, air filter efficiency rating and conditions, and the HVAC maintenance schedule. In addition, general information on merchandise, building materials, store layout, and store activities (e.g., cooking and cleaning) that might influence the indoor contaminants found was also recorded.

The experimental setup in each store was largely the same. Indoor samples of multiple contaminants were collected from a central location on the sales floor. In larger stores, this is supplemented by two to three additional locations on the sales floor where additional VOC samples were collected. Outdoor samples were collected near the main point of entry of outdoor air. For the mechanically ventilated stores, the outdoor location was near a rooftop air intake. For stores that are naturally ventilated, outdoor sampling was conducted near the front doors, as these doors were often left open or were opened and closed frequently. In addition to the sampling of VOCs, other pollutants and indoor environmental metrics were monitored in real-time, including particulate matter (PM), ozone (O_3), carbon monoxide (CO), carbon dioxide (CO_2), temperature, and relative humidity.

3.1 Ventilation Rate

Store ventilation rate was measured using a tracer gas decay method. Sulfur hexafluoride (SF_6) was released in the store, or was injected into the air-handlers, in the early afternoon. Up to four Miran SapphIRe® Model 250B infrared gas analyzers were placed at various locations to monitor the tracer concentrations in real time. A different amount of tracer gas was released depending on the store size to achieve an initial concentration of about 1,000 ppm. In the mechanically ventilated stores, tracer gas concentrations typically become well-mixed in about 30 minutes. This was determined by multiple Miran analyzers reading within 10% of one another. In larger stores (about 1,000 m^2 in floor area and greater) that are naturally ventilated, a fan was used to help with mixing of the tracer gas. Besides the Miran analyzers, grab samples of indoor air

containing SF_6 were also collected in polyethylene-lined multi-layer sample bags at a few locations inside the store. About ten samples were collected over one hour to obtain a range of SF_6 concentrations for comparison with the real-time data. Bag samples were analysed by gas chromatograph equipped with an electron capture detector. Concentrations from the bag samples were compared with Miran measurements as a data check. Some bag samples were also collected at locations outside the sales floor to see if there was air exchange between these spaces. Examples of such spaces include offices, employee break rooms, and warehouse areas.

Air exchange rate was computed from curve fitting to the exponential decay in SF_6 concentrations. The SF_6 concentrations were log-transformed and then plotted against time. Best-fitted slope of the least-square linear regression gives the air exchange rate. This linear regression method was applied to each set of the SF_6 concentrations measured by the Miran analyzers. In addition, the age of air was computed according to ASHRAE Standard 129 (ASHRAE, 2002). The age-of-air approach calculates the average amount of time that has elapsed since a given volume of air entered the building from outside. Standard 129 applies to spaces that are naturally ventilated only in theory, because part of its purpose is to estimate the air-change effectiveness of the mechanical system in terms of mixing and the uniformity of air distribution within the building. However, as long as the tracer gas decay started with a fairly uniform concentration inside the building, the age-of-air method can be applied to all buildings regardless of their ventilation mode.

3.2 Volatile Organic Compounds

VOCs were collected using multi-bed Carbopack X® and Carbopack B® sorbent tubes. One-hour samples were collected in the afternoon at one or more indoor locations, and concurrently at one outdoor location. The sample flow of 0.1 Lpm was monitored continuously using a mass airflow sensor (Series AWM5000 Microbridge; Honeywell) with an estimated measurement error of 3%. The sample flow rates were also verified using a primary airflow calibrator (Gilibrator-2; Sensidyne) with a precision greater than 2% before sampling. Tubes were pre-conditioned in a tube conditioning unit (TCU; Gerstel) with high purity helium at 40 mL/min and 315 ˚C for 1 hour. A duplicate set of samples was collected. The VOC concentrations presented here, including formaldehyde and acrolein that were sampled using a different method as described below, are the averaged values from two co-located sample duplicates.

VOCs were analyzed following U.S. EPA Methods TO-1 and TO-17 (U.S.EPA, 1984, U.S.EPA, 1999). Multi-point internal standard calibrations were performed using pure compounds and 1-bromo-3-fluorobenzene as the reference compound. Tubes were thermally desorbed for analysis by gas chromatography/mass spectrometry (TD-GC/MS) using a thermodesorption auto-sampler (Model TDSA2; Gerstel), a thermodesorption oven (Model TDS3; Gerstel) and a cooled injection system (Model CIS4; Gerstel). Compounds were resolved on a GC (Series 6890Plus; Agilent Technologies) equipped with a 30 meter HP-1701 14% Cyanopropyl Phenyl Methyl column (Model 19091U-233; Agilent Technologies). The resolved analytes were detected using a mass spectrometer (Model 5973; Agilent Technologies). The MS was operated in scan mode. Multipoint calibrations were prepared from pure standards for all target VOCs. All analytes were referenced to the internal standard.

Volatile carbonyl samples were collected using dinitrophenyl hydrazine (DNPH)-coated Waters Sep-Pak® cartridges. Ambient ozone was removed with potassium iodide scrubbers preceding each DNPH samples collected outdoors. One-hour samples were collected at the same indoor and outdoor locations at 1 Lpm. DNPH cartridges were extracted with 2-mL aliquots of acetonitrile, and the extracts were analyzed by high-performance liquid chromatography (HPLC) with UV detection at λ_{max} = 360 nm (Series 1200; Agilent Technologies). A calibration curve for quantification was carried out using standards of the formaldehyde-DNPH hydrazone.

In addition, acrolein samples were collected using pentafluorophenyl hydrazine (PFPH)-coated cartridges that were prepared in our laboratory. This derivatization method is based on Ho and Yu (2004). This is the first field study that used this method of sampling for acrolein. More details about the preparation of the PFPH cartridge and analysis are discussed in later part of this report.

3.3 Particulate Matter

Particle counts and mass concentrations were measured at the same central indoor location and also at the same outdoor location as the VOC samplers. Real-time particle counts were monitored for six to eight hours using an optical particle counter (Model BT-637; MetOne) that measured particle counts at these size bins: >0.3, >0.5, >0.7, >1, >2, and >5 μm. This instrument has a counting efficiency of about 50% for 0.3 μm particles, so particle counts in the first channel are uncertain. It measures up to approximately 100,000 particles/L. The reported measurement accuracy is 10%. Particle counts were measured at one-minute averaging time interval.

Ultrafine particle (UFP) counts were measured at the central indoor location and also outdoors using a water-based condensation particle counter (WCPC Model 3781; TSI). The WCPC counts particles greater than 6 nm. The sample inlet has a cyclone with a cut-off diameter of 3 μm. The instrument measures up to 50,000 particles/cm^3. The reported measurement accuracy is 10%. UFP counts were recorded at an averaging time step of one minute.

Prior to each sampling visit, flow rate of the optical particle counters and the WCPCs were measured and adjusted if needed to meet the operating specifications. A HEPA filter was attached to the sampling inlet to check that the instrument was leak free. Side-by-side comparisons show that the outdoor and indoor MetOne particle counters agreed to within 5% in the four smaller size bins, and agreed within 10% in the two larger size bins. The agreement was worse for the larger particles likely because fewer counts were measured. The two WCPCs used in this study agreed with each other to about 3% prior to sampling. When the instruments were compared side-by-side again after each sampling, they agreed to about 12%. On the other hand, the MetOne particle counters generally maintained the same level of agreement among instruments before and after sampling.

PM2.5 and PM10 samples were collected using a single-stage impactor with a 2.5 and 10 μm cut-point (Model 200 Personal Environment Monitors; SKC). Samples were collected at one outdoor and one indoor location at 10 Lpm for approximately 6 hours. Particle mass was collected onto polytetrafluoroethylene (PTFE) membrane filters (37-mm, 2-μm pore size Teflo

membranes, Pall Life Sciences). Filters were analyzed gravimetrically. The manufacturer assembly procedure was followed in the loading and unloading of the filters. Filters were weighed inside an enclosure where the temperature was maintained between 22 and 26 ^{o}C and relative humidity between 30 and 40%, respectively. Filters were conditioned in this enclosure for 24 hours before each weighing before and after sampling. The balance used for filter weighing has a readability of 0.1 μg (Model CCE6; Sartorius). The accuracy of the balance was checked before each weighing using a NIST certified 0.2 g standard weight. Two field blanks were collected on each sampling day. Measurement blanks averaged to 1.5 μg (standard deviation = 2.7 μg), or approximately 0.4 μg/m^3 for a 6-hour sample. Blanks were subtracted from the weight of the filter samples. The sample flow rates were measured using the same mass flow sensors as those used for the VOC samplers. Prior to each site visit, sample flow rates were measured using a primary airflow calibrator (Gilibrator-2; Sensidyne) and adjusted to within 5% of the specified value as stated in the operation procedure.

3.4 Ozone

Concentrations of O_3 were monitored using real-time gas analyzers (Model 205, 2BTechnologies) at the central indoor location and also outdoors. This dual-beam instrument measures the UV light intensity of ozone-scrubbed and unscrubbed sample air for baseline stability and fast response. The limit of detection is 2 ppb. It has a reported accuracy of 2%. An averaging time of one minute was used in this study. The gas analyzers were checked to make sure that the indoor and outdoor units agreed with one another before and after sampling. Zero-offsets of the instruments were determined in the laboratory by sampling with an O_3 scrubber attached to the sample inlet. The offset values typically ranged between 1 and 5 ppb were subtracted from the field data.

3.5 Carbon Dioxide and Carbon Monoxide

CO_2 and CO were measured at the central indoor location and also outdoor using real-time gas analyzer. The EGM-4 (PP Systems) is a high precision CO_2 analyzer with a reported accuracy of 1%. The gas analyzers used were calibrated for use between 250 and 2,500 ppm using a standard gas as the reference. The soda lime used by the gas analyzers in the absorber column for auto-zeroing was changed as needed per the operation manual. The EGM-4 was used to measured CO_2 concentrations at one-minute time interval.

The Langan Model T15n uses an electrochemical sensor to measure CO concentrations passively. The gas analyzers were calibrated for use between 4 and 180 ppm using a standard gas as the reference. The T15n has a reporting limit of 0.1 ppm. The CO concentrations in retail stores, measured at one-time averaging time, were often at or below the reporting limit.

3.6 Temperature and Relative Humidity

The indoor air temperature and relative humidity were monitored at multiple locations within the stores using HOBO U12 temperature/relative humidity data loggers. The data loggers also functioned as data storage devices for sample flows and concentration measurements from other instruments. The outdoor air temperature and relative humidity were recorded by a HOBO that was co-located with the suite of outdoor sampling instruments.

4 Store Descriptions

Nineteen retail stores located in various California cities were sampled (Table 1); see Figure 1 for the location of stores in relation to California climate zones. Most of the stores were opened or renovated within the past 15 years. The sample included seven grocery stores (G1–G8, with G7 being a second visit to G3), seven furniture/hardware stores (F1–F8, with F7 being a second visit to F6), and five apparel stores (A1–A5).

Table 1 Building characteristics of nineteen retail stores.

	City	Store Open	Sample Date(s)	Floor Area (m^2)	Ceiling Height (m)	Max # People in Store	Ventilation Mode	Air Filter MERV Rating
Grocery Stores								
G1	Berkeley	2009	2011-9-27 & 28	3,270	7.5	260	Mech.	15
G2	Walnut Creek	2001	2011-10-6 & 7	1,840	6.8	180	Mech.	8
G3	Tarzana	2010	2012-5-14	3,310	10.4	120	Mech.	8
G4	W. Sacramento	2006	2012-9-5	3,720	7.0	90	Mech.	8
G5	Roseville	2007	2012-9-6	3,720	6.9	60	Mech.	8
G6	Brea	2011	2013-2-11	1,240	8.7	65	Mech.	2
G7	See G3 (revisit)		2013-2-28	See G3		130	See G3	
G8	Fairfax	2012	2013-3-26	1,310	5.8	130	Mech.	8
Furniture/ Hardware Stores								
F1	San Francisco	2009	2011-10-24 & 25	640	4.0	5	Nat.	N/A
F2	Oakland	2004	2011-11-1	1,530	6.0	10	Nat.	N/A
F3	Berkeley	1995	2011-11-2	680	4.0	8	Nat. (Mech.)	Unknown
F4	Fremont	2011	2012-6-1	5,110	7.5	70	Mech.	8
F5	Long Beach	2012	2012-6-5	790	3.6	10	Nat. (Mech.)	2
F6	Fresno	2008	2012-9-10	3,710	6.3	60	Mech.	2
F7	See F6 (revisit)		2013-2-13	See F6		65	See F6	
F8	Costa Mesa	1980s	2013-2-27	1,020	5.1	25	Mech.	2
Apparel Stores								
A1	Oakland	1997	2012-2-21	120	3.1	10	Nat. (Mech.)	8
A2	Oakland	2011	2012-2-22	80	3.4	5	Nat. (Mech.)	Unknown
A3	San Mateo	2011	2012-2-28	1,070	9.3	25	Mech.	8
A4	Long Beach	2011	2012-6-6	990	5.3	40	Mech.	8
A5	Folsom	2012	2013-3-27	130	6.8	5	Mech.	2

Information about the research study was sent to store owners and managers by mail and via email to recruit their stores to participate. Some of the stores located nearby were also visited in person during recruiting. Because recruiting was challenging, enrollment of the stores formed a convenient sample rather than trying to be statistically representative. Stores were offered a monetary incentive for their assistance during our sampling. Each participating store also received a data report that described the measured ventilation rate and indoor air quality, and how the measurements compared to recommended levels. The study protocol was reviewed and approved by Lawrence Berkeley National Laboratory's institutional review board. The storeowner or manager provided informed consent.

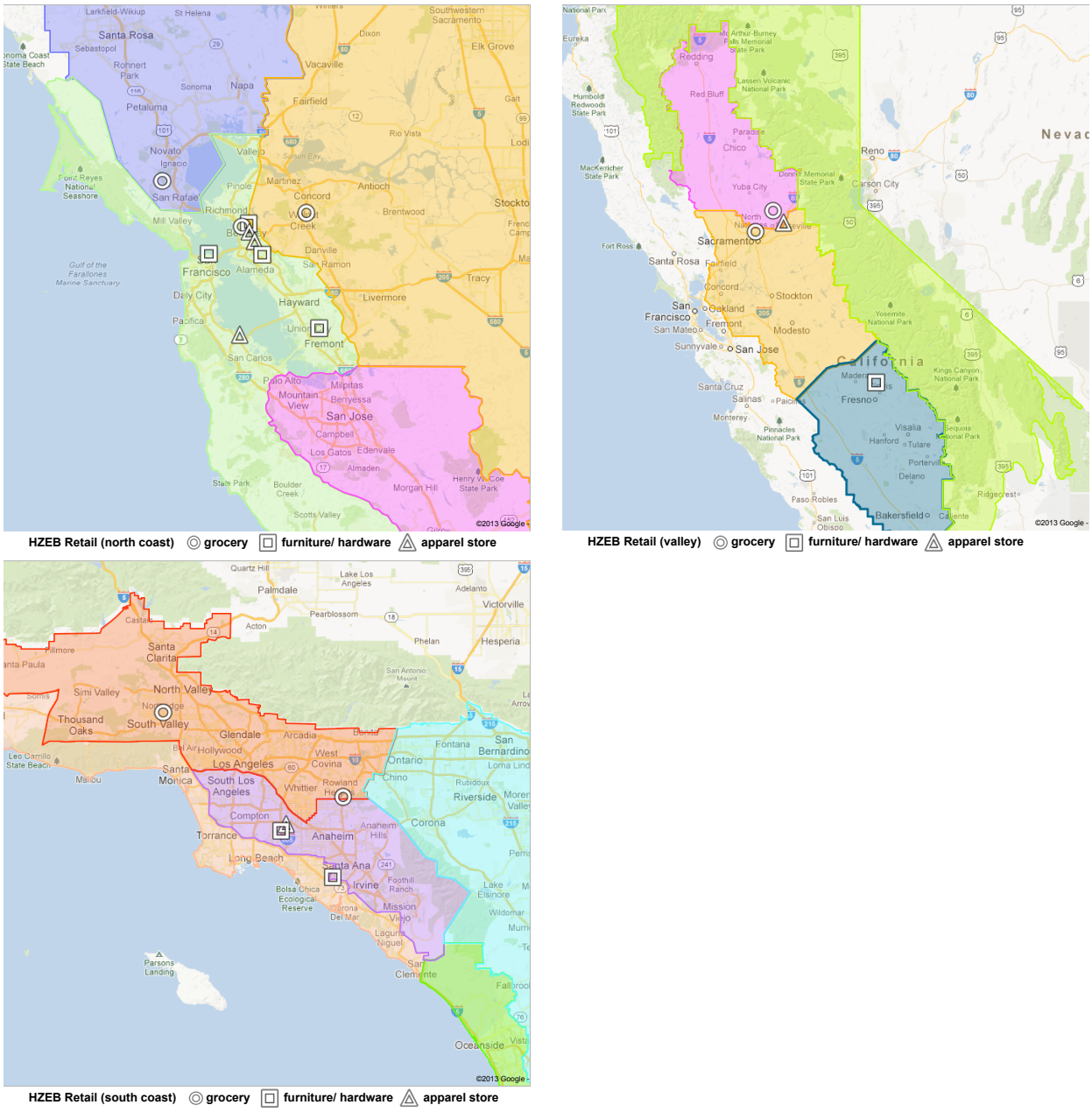

Figure 1 Map showing locations of the retail stores sampled in north coast, central valley, and south coast of California (climate zones are shown as background).

With the exception of F1 and F2, all other stores had rooftop heating, ventilating, and air conditioning (HVAC) units to condition the indoor spaces. However, smaller stores, such as F3, F5, A1, and A2, preferred to rely on natural ventilation for most of the year. On the sampling days, these stores operated as usual i.e., the data for six stores (F1, F2, F3, F5, A1, and A2) was collected when they were naturally ventilated. In stores F5 and A1, ventilation rates were measured twice, first with the front door opened and HVAC off. During this operate-as-usual period, contaminant concentrations were measured. Following this, the ventilation rate was measured for a second time with HVAC on and the front door closed.

With the exception of two stores where the rooftop HVAC was inaccessible, the particle removal efficiency, i.e. MERV ratings, of the air filters were noted. The majority of the stores (nine) have MERV 8 air filters. Five stores have MERV 2 air filters. The highest efficiency air filters (MERV 15) were used at one grocery store that was built in 2009 (Table 1).

The sampled grocery stores tended to be the largest in floor area and also had the most number of occupants. Apparel stores tended to be the smallest. Table 1 gives the total floor area of the retail stores, which included not only the sales floor, but also offices, warehouses, employee areas, and other spaces within the stores. This definition was used because it is the most commonly known floor area to the storeowner or manager. Occupancy varied greatly in a store as a function of the time of day. Table 1 shows the maximum number of occupants counted, including all employees and shoppers. Because all stores were sampled during a weekday, these people counts do not reflect the maximum occupancy that can be expected on weekends and holidays.

5 Results

5.1 Ventilation Rate

Table 2 shows all the ventilation rate measurements collected using the SF_6 decay method. In most cases, the decay time constants resulted from the slopes of the SF_6 measured by the different Miran analyzers agreed with one another within about 10%. The estimated air exchange rates on two days were within 10% for the two mechanically ventilated stores G1 and G2. But substantial spatial and temporal differences in air exchange rates were observed in some of the naturally ventilated stores. In store F1 for example, which was naturally ventilated, the difference between the two days was 20%. In store F3, the three Miran analyzers agreed initially by using a fan to obtain a uniform SF_6 concentration. But once the fan was turned off, SF_6 concentrations varied spatially in the store as a result of poor mixing of the indoor air. In store A2, air exchange rate varied substantially during the day between 1 and 4 h^{-1}. The air exchange rates shown in Table 2 are the averages estimated from multiple curve fittings to portions of SF_6 during which the decay rates were roughly linear on log-scale.

Note also in Table 2, the ventilation rates of stores A1 and F5 were measured twice, first as a naturally ventilated store, and later as a mechanical ventilated store with the HVAC on. In both stores, the ventilation rate with HVAC off and front door opened was substantially higher than with HVAC on and front door closed.

In addition to estimating the air exchange rates from the slopes of the SF_6 decay curves, the mean age of air was also calculated following Standard 129 (ASHRAE, 2002). The rightmost column in Table 2 shows the air exchange rates computed from the inverse of the mean age of air. These estimates are also plotted Figure 2. Overall, there is a closer agreement between the age of air estimated from the different Mirans, compared to the time constants estimated from the slopes of the SF_6 decay curves. Imperfect mixing is the key cause of differences in the ventilation rate estimates from the different Mirans. The two methods of estimating the air exchange rates, (i) fitting the slope of decay curve and (ii) calculating the age of air, gave results that are highly

correlated ($R^2 = 0.92$). In general, method (i) tended to give air exchange rate estimates that are higher than method (ii).

Table 2 Ventilation rates estimated using the tracer gas decay method.

Stores	# Mirans	Decay Time Constant (hours^{-1})			Age of Air (hours)			ACH = (Age of Air)$^{-1}$
		Mean	Range	% Difference	Mean	Range	% Difference	
Grocery Stores								
G1 (1) *	3	0.64	0.61–0.67	10%	1.36	1.34–1.41	5%	0.74
G1 (2) *	3	0.72	0.69–0.79	13%	1.30	1.26–1.33	5%	0.77
G2 (1) *	3	1.98	1.84–2.06	11%	0.68	0.63–0.76	19%	1.47
G2 (2) *	3	1.78	1.70–1.84	8%	0.70	0.66–0.77	16%	1.43
G3	4	0.70	0.66–0.73	10%	1.30	1.22–1.39	13%	0.77
G4	4	0.76	0.66–0.81	20%	1.34	1.12–1.52	30%	0.75
G5	4	0.51	0.42–0.56	27%	1.53	1.37–1.72	23%	0.65
G6	4	0.98	0.92–1.05	13%	0.89	0.87–0.91	4%	1.12
G7	4	0.72	0.65–0.84	27%	1.31	1.20–1.45	19%	0.65
G8	4	2.70	2.64–2.78	5%	0.50	0.49–0.51	4%	2.00
Furniture/Hardware Stores								
F1 (1) *	3	1.26	1.20–1.32	10%	0.81	0.74–0.87	16%	1.23
F1 (2) *	3	1.02	0.98–1.05	7%	0.99	0.88–1.06	18%	1.01
F2	3	2.84	2.63–3.03	14%	0.42	0.36–0.46	24%	2.38
F3	3	0.35	0.20–0.61	118%	2.59	1.97–3.06	42%	0.39
F4	3	0.64	0.59–0.68	13%	1.47	1.34–1.58	16%	0.68
F5 (N) **	2	2.28	2.21–2.35	6%	0.86	0.80–0.91	13%	1.16
F5 (M) **	2	0.98	0.75–1.21	46%	1.10	1.04–1.16	11%	0.91
F6	4	0.45	0.44–0.46	5%	1.90	1.86–1.96	5%	0.53
F7	4	0.47	0.44–0.53	19%	1.38	1.22–1.53	22%	0.72
F8	3	1.62	1.04–2.01	60%	0.86	0.74–1.07	38%	1.16
Apparel Stores								
A1 (N) **	2	3.86	3.42–4.29	23%	0.43	0.42–0.44	5%	2.33
A1 (M) **	1	1.08	--	--	1.00	--	--	1.00
A2	1	3.04	--	--	0.45	--	--	2.22
A3	3	0.41	0.39–0.44	11%	1.94	1.90–1.98	4%	0.52
A4	2	0.46	0.45–0.47	5%	1.86	1.74–1.97	12%	0.54
A5	1	2.61	--	--	0.46	--	--	2.19

* For stores where the ventilation rates were measured more than once: (1) and (2) refers to day one and two of sampling;
** (N) refers to natural ventilation, and (M) refers to mechanical ventilation.

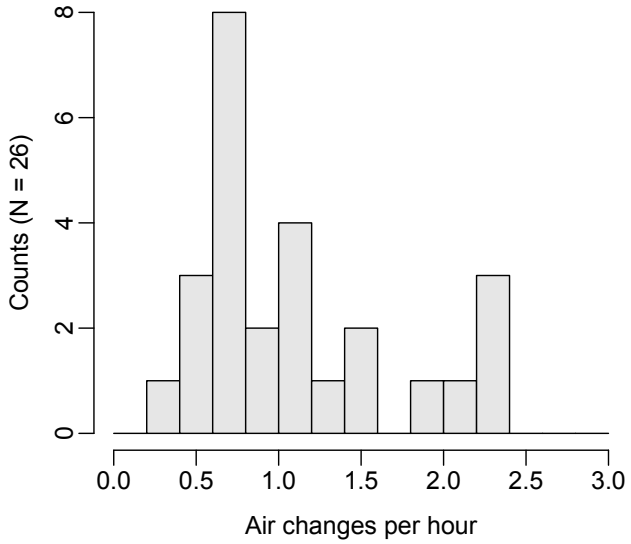

Figure 2 Air changes per hour estimated from SF_6 decay from 21 store visits. Three stores were measured on two days, and two stores were measured once with HVAC off, and once with HVAC operating (see Table 2). Thus, a total of 26 estimates are plotted. The air changes per hour was computed using the inverse of the mean age of air.

California's Title 24 standard requires that all stores met the minimum ventilation rate on a per person basis (7 L/s-person) and also on a per floor area basis (1 L/s-m^2). Figure 3 shows the estimated ventilation rates compared with Title 24 at these two reference values. All stores met the per floor area requirement of 1 L/s-m^2, with the exception of F3. F3 had low ventilation rate because its front door was closed on the day of sampling, and there were no other intentional openings that would allow outdoor air to enter the store.

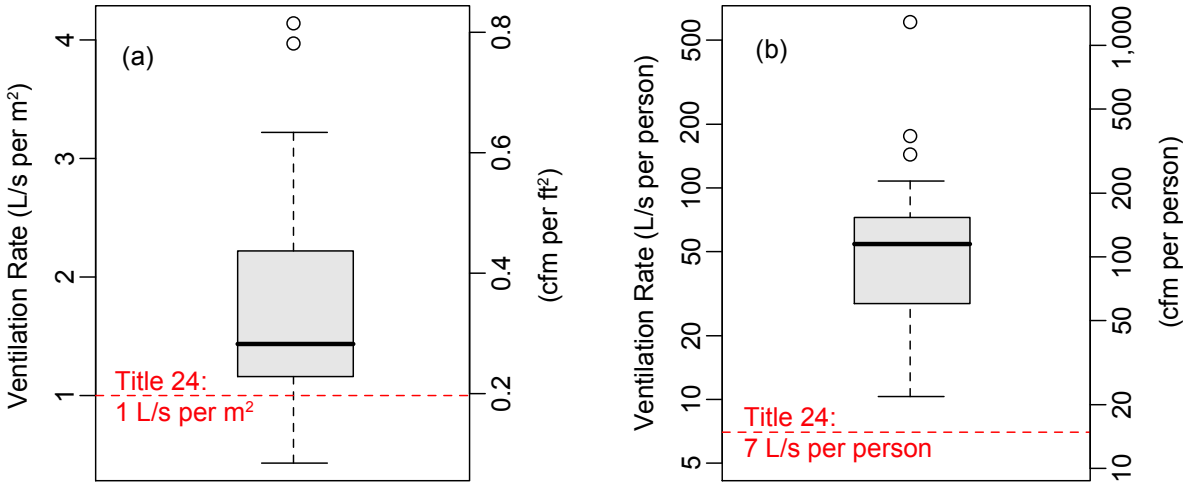

Figure 3 Ventilation rates estimated from 21 store visits on (a) per person and (b) per floor area basis. The red lines indicate California's Title 24 standards.

Furniture/hardware stores had the highest ventilation rate estimates on a per person basis because they generally had fewer occupants per floor area. But this can change over time because occupancy can vary greatly in retail stores. For example, store employees in several apparel stores indicated that the number of customers can increase by five folds on weekends and holidays. In grocery stores, the number of customers can increase by a factor of two or three in the evenings and on weekends. This would imply a ventilation rate closer to the minimum requirements during busy hours.

Table 3 Comparison of measured air changes per hour and Title 24 requirements.

Grocery Stores			**Furniture/Hardware Stores**			**Apparel Stores**		
	Measured ACH	Title 24 ACH		Measured ACH	Title 24 ACH		Measured ACH	Title 24 ACH
G1	0.75	0.48	F1 (N)*	1.12	0.90	A1 (N)*	2.33	1.16
G2	1.45	0.53	F2 (N)*	2.38	0.60	A2 (N)*	2.22	1.06
G3 / G7	0.71	0.35	F3 (N)*	0.39	0.90	A3	0.52	0.39
G4	0.75	0.51	F4	0.68	0.48	A4	0.54	0.68
G5	0.65	0.52	F5 (N)*	1.16	1.00	A5	2.19	0.53
G6	1.12	0.41	F6 / F7	0.63	0.57			
G8	2.00	0.62	F8	1.16	0.70			
Average =	1.06	0.49	Average =	1.07	0.74	Average =	1.56	0.76

* (N) denotes stores that relied on natural ventilation as their dominant mode.

Another way to compare the measured ventilation rates with California's minimum ventilation rate requirement is to compute a Title 24-equivalent air changes per hours. In Eq 1, the per floor area ventilation rate of 1 L/s per m^2 is divided by the store height H (m), and is multiplied by a factor of 3.6 for unit conversion.

$$\mathrm{ACH}_{\text{Title 24}} = 1\frac{\mathrm{L}}{\mathrm{s}\cdot\mathrm{m}^2} \times \frac{3.6}{H(\mathrm{m})}$$

Eq 1

Table 3 shows that on average, stores were adequately ventilated when compared with Title 24 in terms of air changes per hour. The average ACH was about 1 h^{-1} in grocery and furniture/hardware stores, and 1.5 h^{-1} in apparel stores. Grocery stores tend to have taller ceilings (average height = 7.6 m), which explain the lower Title 24-equivalent ACH than the other two store type types (average height = 5.2 m and 5.6 m, respectively). This is important because store heights vary greatly, from 3 m (store A1) to 10 m (store G3) in this study for example. If the source strengths of air contaminants scale with floor area, then stores with the same ventilation rate per unit floor area would have the same indoor contaminant concentrations, regardless of their air exchange rates being different because of store height. However, if contaminant source strengths scale with the indoor air volume, then contaminant concentrations will depend on air exchange rates.

The median air changes per hour was about 0.75 h^{-1} in stores that were ventilated mechanically, as shown in Figure 4. In comparison, stores that relied on natural ventilation either exclusively or most of the time had higher ventilation rates than mechanically ventilated stores. Store workers expressed that they preferred the sense of more air being moved through the store when doors are

left opened to allow for natural ventilation. The decision to switch to mechanical ventilation was largely based on thermal comfort inside the store. If heating or air conditioning was needed to adjust the indoor temperature, then the stores would close the doors and turn on the HVAC. There are three mechanically ventilated stores with air exchange rates above 1.5 h^{-1}: G2, G8, and A5. These very high ventilation rates were observed likely because the stores were operating in economizer mode. All three stores were located in the coastal region of northern California, where the climate is moderate. On the days of sampling, the outside air temperature was at least partly in the 10 to 20 ^{o}C range. Buildings equipped with economizers would bring in more outside air for cooling, which would explain these high air exchange rates.

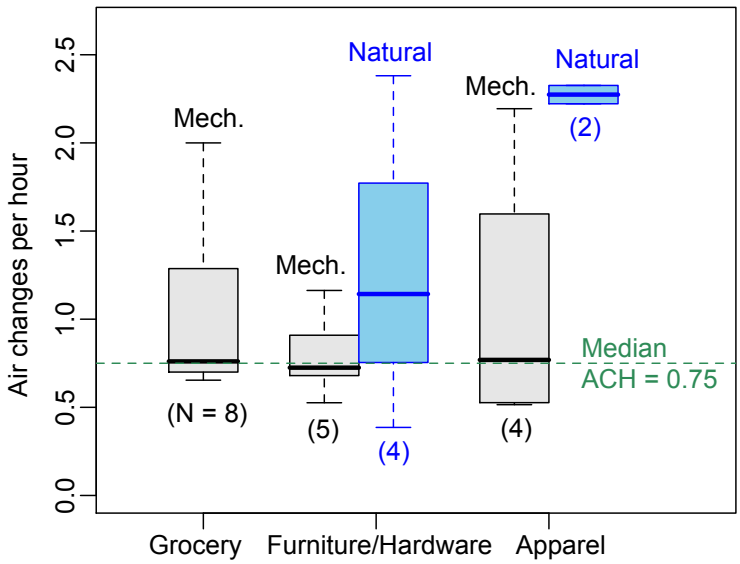

Figure 4 Measured ventilation rates for three store types, plotted separated by their dominant ventilation mode. N is the number of store visits represented in each boxplot.

5.2 Carbon Dioxide

Carbon dioxide concentrations were measured for 6 to 7 hours both indoors and outdoors simultaneously. The monitoring hours were generally between 10 am and 5 pm. All the stores maintained low level of CO_2 concentrations because of the relatively high per person ventilation rates, as shown in Figure 3. ASHRAE Standard 62.1 (2010) originally linked the minimum ventilation rates to maintaining the indoor CO_2 concentration to no greater than 700 ppm above the outdoor air levels. This criterion is met in all the stores by a large margin on the day of the sampling.

Figure 5 shows that the difference between the 1-hour maximum indoor CO_2 and the average outdoor level typically ranges between 100 and 400 ppm. In the three stores (G1, G2, and F1) where monitoring continued for two days, the day-to-day variability in the indoor CO_2 concentrations is about 20%. However, store occupancy can increase substantially during peak hours and on busy days such as weekends and holidays. If the number of people in a store were

to increase by a factor of two, so would the difference between indoor and outdoor CO_2 concentration, assuming that the ventilation rate of the stores was unchanged.

As expected, Figure 5 shows that there is an inverse correlation between indoor CO_2 concentrations and the per person ventilation rate measured. In an ideal case where the occupancy and ventilation rate were held constant throughout the day, the per-person ventilation rate would determine the indoor CO_2 concentrations. However, the absence of tighter correlation in Figure 5 is likely the result of variation in occupancy and ventilation rates during the measurements.

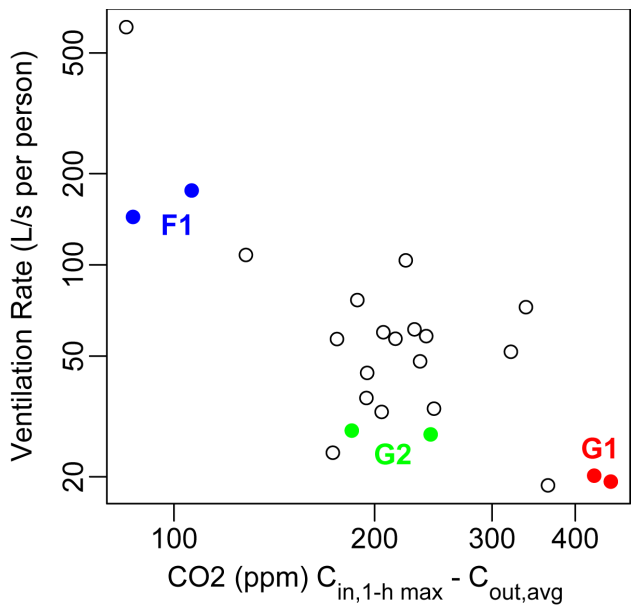

Figure 5 Relationship between the 1-hour maximum indoor CO_2 (x-axis) and store per-person ventilation rate (y-axis) from 21 store visits (note: both axes shown in log-scale). The average outdoor CO_2 was subtracted from the indoor values. The per-person ventilation rates were computed using the maximum occupancy observed on the day of sampling. Three stores were sampled on two days instead of one, as highlighted in color.

The measured CO_2 and ventilation rate data allow rough estimates of the CO_2 generation rate in retail stores, as follows:

$$G = \frac{(C_{in} - C_{out}) \cdot Q}{N}$$

Eq 2

where G (L/s-person) is the CO_2 generation rate per person, C_{in} and C_{out} (ppm) are the indoor and outdoor CO_2 concentrations, Q (m^3/h) is the outdoor air flow rate, and N is the number of people in the store. G is a useful estimate for modeling the ventilation need of occupants in retail stores. Eq 2 assumes that all variables are constants, and steady state has been reached. Even though these conditions were not met during our store visits, reasonable estimates of G could be made by setting C_{in} to equal the 1-hour maximum indoor CO_2 concentrations, as shown in Figure 5,

and the per-person ventilation rate Q/N to equal the values shown on the y-axis. Using these approximations, the CO_2 generation rates estimated have a median value of 0.012 L/s.

Emmerich and Persily (2001) summarized the literature on demand controlled ventilation based on CO_2 level in buildings, and estimated 0.005 L/s for an average adult engaged in office work. The rough estimate of 0.012 L/s for retail stores calculated here corresponds to a physical metabolic activity of approximately 2.5, which is similar to walking at a pace of 2 miles per hour or house cleaning (ASHRAE, 1997). Anecdotally, more heavy lifting was observed in furniture/hardware stores. This would explain the median CO_2 generation rate was also the highest in furniture/hardware stores (G = 0.014 L/s) compare to the other store types. Grocery and apparel stores had lower estimates of median CO_2 generation rate: G = 0.010 and 0.0082 L/s, respectively. Workers in those stores performed activities such as shelving, helping customers with merchandises and working at check-outs. The lower G estimates would suggest that these work duties may be lower in physical metabolic activity than those in furniture/hardware stores.

Table 4 Measurements of carbon dioxide and carbon monoxide.

	Sampling Time		Carbon Dioxide (ppm)			Carbon Monoxide (ppm)		
	Start	**End**	**Outdoor Average**	**Indoor Average**	**Indoor 1-h Max**	**Outdoor Average**	**Indoor Average**	**Indoor 1-h Max**
Grocery Stores								
G1 (1) *	10:40	16:40	381	777	833	0.6	1.4	1.5
G1 (2) *	09:35	16:20	405	768	832	0.9	1.6	1.8
G2 (1) *	10:40	17:55	363	536	548	0.3	1.2	1.3
G2 (2) *	08:05	16:10	370	570	613	1.1	1.4	1.7
G3	09:30	17:05	370	581	599	0.7	2.1	2.2
G4	10:00	17:25	400	575	607	1.3	1.9	4.3
G5	10:30	17:10	346	577	585	0.6	1.1	1.6
G6	10:05	17:05	350	638	670	1.0	1.2	1.3
G7	11:15	17:20	348	564	582	0.1	1.3	1.3
G8	11:10	17:20	406	579	611	0.3	1.3	1.3
Furniture/ Hardware Stores								
F1 (1) *	12:10	17:55	345	417	452	0.3	1.4	1.5
F1 (2) *	11:30	16:40	353	417	440	0.4	1.4	1.6
F2	10:55	17:00	350	422	435	1.0	1.0	1.2
F3	10:40	17:05	363	538	557	0.6	1.3	1.3
F4	10:25	17:55	355	552	578	0.8	1.3	1.4
F5	09:55	17:55	389	523	577	0.8	1.3	1.4
F6	10:45	17:10	388	587	603	0.7	0.5	0.6
F7	10:25	17:00	346	597	683	0.5	1.3	1.6
F8	10:35	17:05	358	503	553	1.3	1.3	2.0
Apparel Stores								
A1	11:15	16:40	391	514	564	1.3	1.1	1.2
A2	12:20	17:35	373	584	618	0.4	0.9	1.1
A3	10:20	17:25	359	510	535	0.2	0.1	0.1
A4	11:55	17:55	356	704	720	0.7	1.1	1.2
A5	11:15	17:55	370	472	498	0.5	1.2	1.2

* Stores G1, G2, and F1 were measured on two days, as indicated by (1) and (2).

5.3 Carbon Monoxide

Carbon monoxide concentrations measured were low both outdoors and indoors. The average concentrations measured were typically in the range of 0.5 and 1 ppm. Grocery stores G3 and G4 had the highest average indoor CO concentration at roughly 2 ppm. In G4, the maximum 1-hour concentration reached 4 ppm in the morning, where emissions from cooking in preparation for lunchtime could be a source of CO. In F8, the maximum 1-hour concentration measured indoors coincided with the peak outdoor CO concentration of 4 ppm. Since this hardware store had multiple opened doorways connecting its warehouse to outside, outdoor CO was likely the source of the indoor CO measured. For the stores studied, carbon monoxide was not a concern.

5.4 Ozone

Table 5 shows the ozone concentrations measured in 17 stores. Data was lost from some of the stores sampled due to operator error. The average outdoor ozone concentrations measured outside of the stores varied greatly from 12 ppb at A1 to nearly 70 ppb at G7. These large differences in outdoor ozone concentrations impacted the ozone concentration indoors. In G7, the average indoor ozone concentration was 39 ppb, whereas in A1, the average concentration was only 5 ppb. The indoor-to-outdoor (I/O) ratios shown in Table 5 were computed using the ozone concentrations averaged over the sampling period typically between 10 am and 5pm.

Table 5 Measurements of ozone concentrations, estimated values of I/O ratio and loss rate.

	Sampling Time		Outdoor Ozone (ppb)		Indoor Ozone (ppb)		I/O Ratio	Loss Rate
	Start	End	Average	1-h Max	Average	1-h Max		k_d (h^{-1})
Grocery Stores								
G2 (1) *	10:40	17:55	24.1	30.0	13.9	16.9	0.58	1.1
G3	09:45	17:00	58.8	71.9	25.5	36.6	0.43	1.0
G4	10:35	17:20	46.0	62.9	12.2	18.8	0.27	2.1
G5	10:30	17:05	55.3	79.4	9.8	16.3	0.18	3.0
G6	10:05	17:05	37.0	40.4	12.0	17.4	0.32	2.3
G7	11:10	17:10	66.7	68.8	39.2	42.1	0.59	0.5
G8	11:20	17:35	34.0	36.3	15.6	17.2	0.46	2.4
Furniture / Hardware Stores								
F1 (N) **	11:40	16:40	30.1	30.1	12.7	14.6	0.47	1.2
F2 (2) (N)	11:00	17:00	38.3	38.3	25.5	35.6	0.73	0.9
F4	10:10	17:55	35.5	35.5	7.0	11.4	0.29	1.7
F5 (N) **	09:55	17:55	53.8	53.8	13.3	21.5	0.32	2.0
F6	10:40	17:10	62.1	62.1	15.5	19.3	0.29	1.3
F7	10:20	17:10	50.3	50.3	15.5	27.6	0.35	1.3
F8	10:20	17:05	43.3	43.3	15.3	23.6	0.46	1.4
Apparel Stores								
A1 (N) **	12:15	17:40	12.1	15.1	5.0	5.3	0.41	1.4
A4	12:25	17:55	51.5	59.6	5.9	7.8	0.11	4.2
A5	13:05	17:55	47.9	53.4	22.6	28.0	0.47	2.5

* Stores G2 and F2 were monitored for two days, but only the data from day 1 at G2, and day 2 at F2, were recorded successfully.

** Stores F1, F2, F5, and A1 were naturally ventilated during sampling.

An indoor-outdoor ratio that is less than one implies that there are losses of O_3, such as when the outside air passes through air filters and other components of the HVAC (Zhao, 2007). In addition, there are other important factors that can influence the ozone concentration indoors (Weschler, 2000), such as losses to indoor surfaces and reactions between ozone and other chemicals in the indoor air. Among the 17 stores, the average I/O ratio is 0.4, with values ranging from 0.1 (A4) to 0.7 (F2).

The first order ozone loss rate can be estimated using the air exchange rate and the I/O ratio, as follows:

$$\text{I/O ratio} = \frac{k}{k_d + k}$$

Eq 3

where k (h^{-1}) is the air exchange rate and k_d (h^{-1}) is the first order ozone loss rate.

Table 5 shows the values of k_d computed using the I/O ratios and Eq 3. The average loss rate is 1.8 h^{-1}, with values typically between 1 and 3 h^{-1}. There may be differences in the surface removal rate by store type, with higher rates estimated in apparel stores (average = 2.7 h^{-1}), and lower rates estimate in furniture/ hardware stores (average = 1.4 h^{-1}). This general tend, though uncertain because of the small sample size, agrees with observations that there are more soft materials in apparel stores that would remove ozone more effectively than hard surfaces in hardware stores. Grocery stores have an average k_d = 1.8 h^{-1}.

Stores that used natural ventilation during sampling, i.e. F1, F2, F5, and A1, had k_d that was lower (average = 1.4 h^{-1}) than stores that used mechanical ventilation (average = 1.9 h^{-1}). The mechanical ventilation system likely contributed to losses in ozone as the outside air was distributed in the building, which would explain this difference in the estimated loss rates.

The highest I/O ratio occurred in a naturally ventilated store (F2). This store had high air exchange rate of 2.4 h^{-1} (Table 2) as outdoor air entered into the stores through open doors. The store also had a low estimate of the ozone loss rate of 0.9 h^{-1} (Table 5). Both of these factors contributed to a higher I/O ratio. The average I/O ratio among the four naturally ventilated stores was 0.48. For the mechanically ventilated stores, the average I/O ratio was 0.37.

Assuming that the loss of ozone from reactions with other indoor air pollutants is negligible, Eq 4 predicts the change in indoor ozone concentration in a well-mixed building, where the changes in concentrations are caused by outdoor ozone entering into the store and losses of ozone to indoor surfaces. Because the outdoor concentrations varied substantially during sampling, a simple way to estimate the indoor ozone concentrations is by computing the values of $C_{in}(t)$ discretely at a time step (Δt) of one minute, as follows:

$$\frac{dC_{in}}{dt} = k\left(C_{out} - C_{in}\right) - k_d C_{in}$$

$$C_{in}(t) = \left[k\left(C_{out,t-1} - C_{in,t-1}\right) - k_d C_{in,t-1}\right]\Delta t + C_{in,t-1}$$

Eq 4

This simplified model predicts the indoor ozone concentrations quite well in grocery stores, as shown in Figure 6. This shows that for these stores, the indoor ozone concentrations can be predicted reasonably well from the outdoor concentrations and the values of k_d estimated in Table 5.

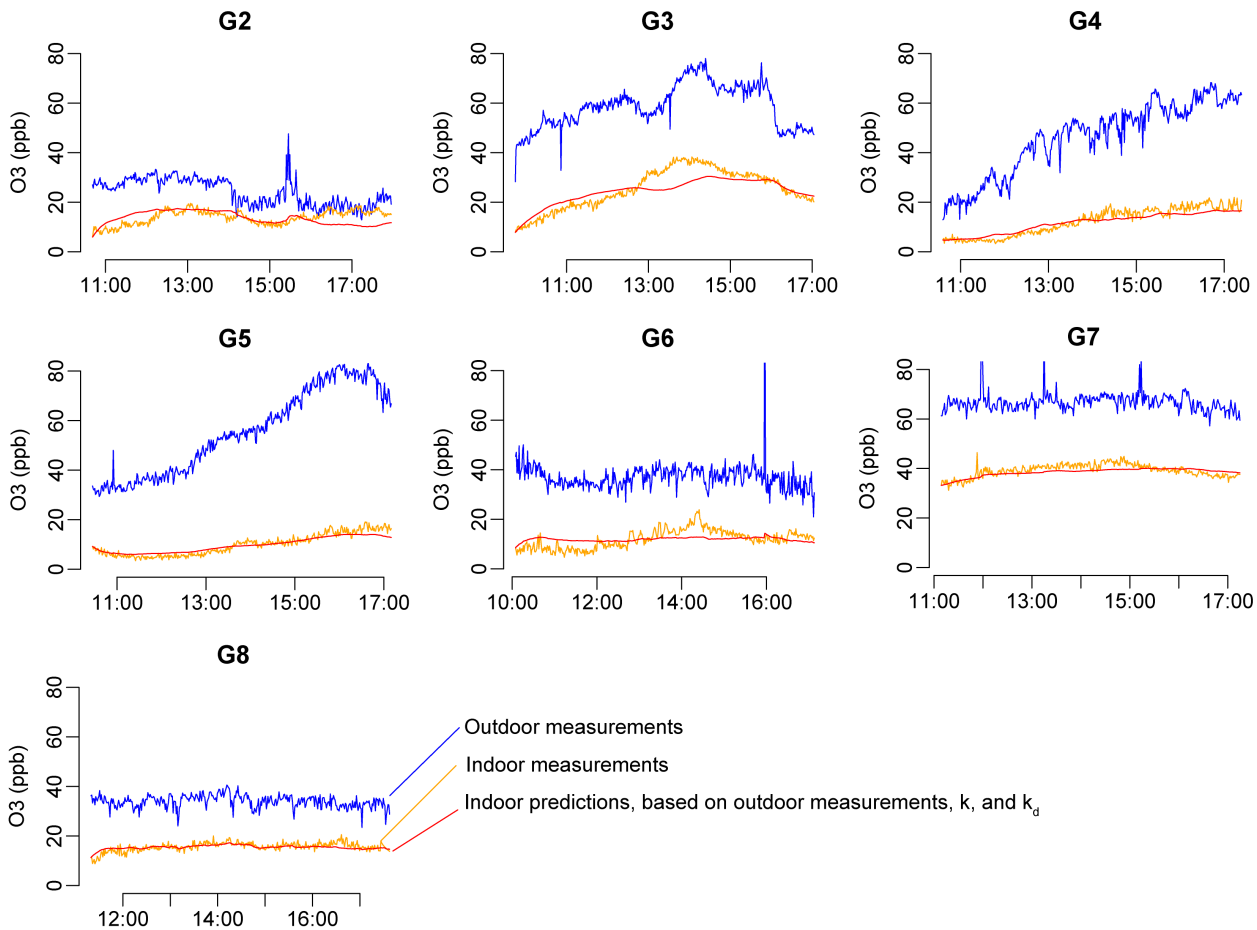

Figure 6 Measurements of outdoor and indoor ozone in seven grocery stores. Predictions of indoor ozone concentrations used air exchange rates estimated in Table 2, and first order loss rates of ozone estimated in Table 5.

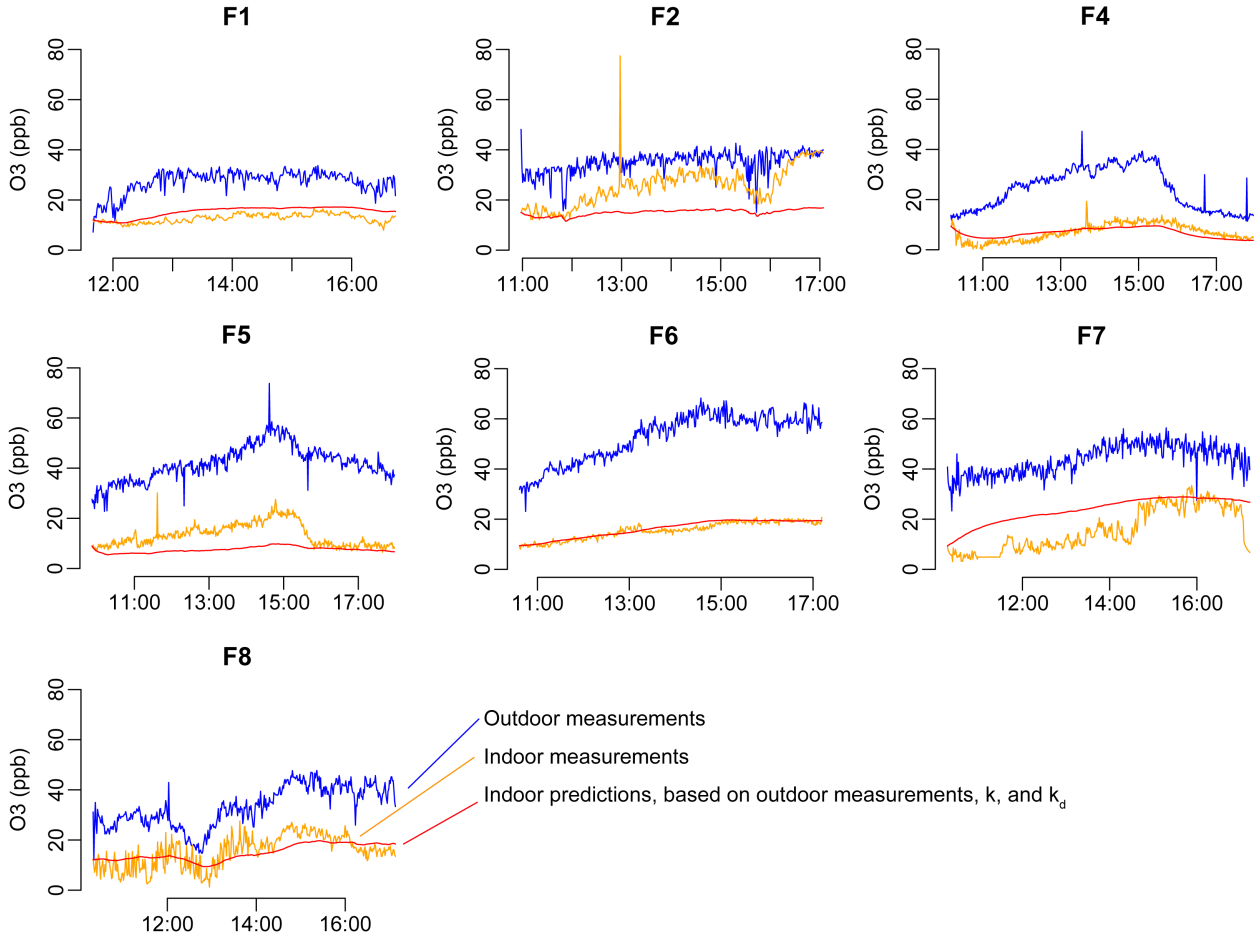

Figure 7 Measurements of outdoor and indoor ozone in two furniture stores (F1 and F2), and in five hardware stores (F4 to F8). Predictions of indoor ozone concentrations used air exchange rates estimated in Table 2, and first order loss rates of ozone estimated in Table 5.

However, in several furniture and hardware stores that used natural ventilation (for example, F2 and F5 in Figure 7), varying rates of air exchange during sampling likely impacted the fit between this simplified model and the measured indoor ozone concentrations. During the first hour of sampling between 11 am and noon, the I/O ratio at F2 was 0.50. At the end of the sampling day between 5 and 6 pm, the I/O ratio increased to 0.93. Increasing air exchange with the outdoors likely caused this dramatic change in I/O ratio of ozone. The reverse is observed in hardware store F5 that also used natural ventilation during sampling, where the I/O ratio decreased from 0.30 to 0.23 on the day of sampling.

In several mechanically ventilated stores, such as F7, F8, and A5, the indoor ozone concentrations varied with time quite rapidly, suggesting that was a change in the air exchange rate with the outdoors. Store F8 is connected to a warehouse that left the rollup door opened to the outside. The flow of outdoor air into the store through the warehouse likely varied during sampling. In F7, indoor temperature controlled operations of the mechanical system. As a result,

many of the rooftop HVAC units were not in use until later in the day when the indoor temperature began to rise. This would explain the increase in the I/O ratio from 0.15 in the morning to 0.52 at the end of sampling. A5 is a small apparel store that is serviced by a single rooftop unit. This unit apparently cycled 9 minutes on and 12 minutes off on the day of sampling. The store had an estimated air exchange rate of 5 h^{-1} when the system was on, which explains the rapid increase in indoor ozone concentration, as shown in Figure 8.

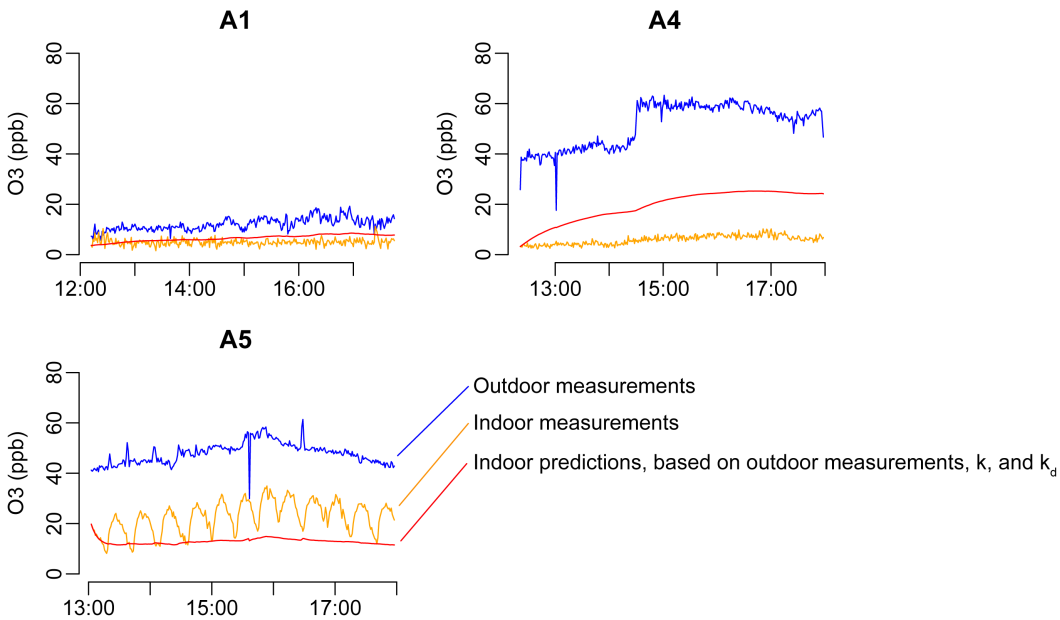

Figure 8 Measurements of outdoor and indoor ozone in three apparel stores. Store A5 showed indoor ozone concentrations that cycled periodically with the rate of the outside air intake. Predictions of indoor ozone concentrations used air exchange rates estimated in Table 2, and first order loss rates of ozone estimated in Table 5.

5.5 Particulate Matter

Figure 9 to Figure 11 shows the average outdoor and indoor particle count measurements by the optical particle counters. Particle counts were measured typically between 10 am and 5 pm for an average duration of 6.5 hours. The particle counters measured the number of particles that are 0.3 μm and larger in six channels, where the last channel included all particles that are 5 μm and larger. In Figure 9 to Figure 11, 10 μm was assumed to be an approximate upper limit of the measurable particle diameter. The y-axis shows the particle count concentrations ΔN (#/L) measured within each of the six particle size bins, normalized to width of the particle size bin Δdp (μm), that gives the particle number distribution $n(dp)$ in units of particle counts (#) per air volume (L) and bin-width (μm).

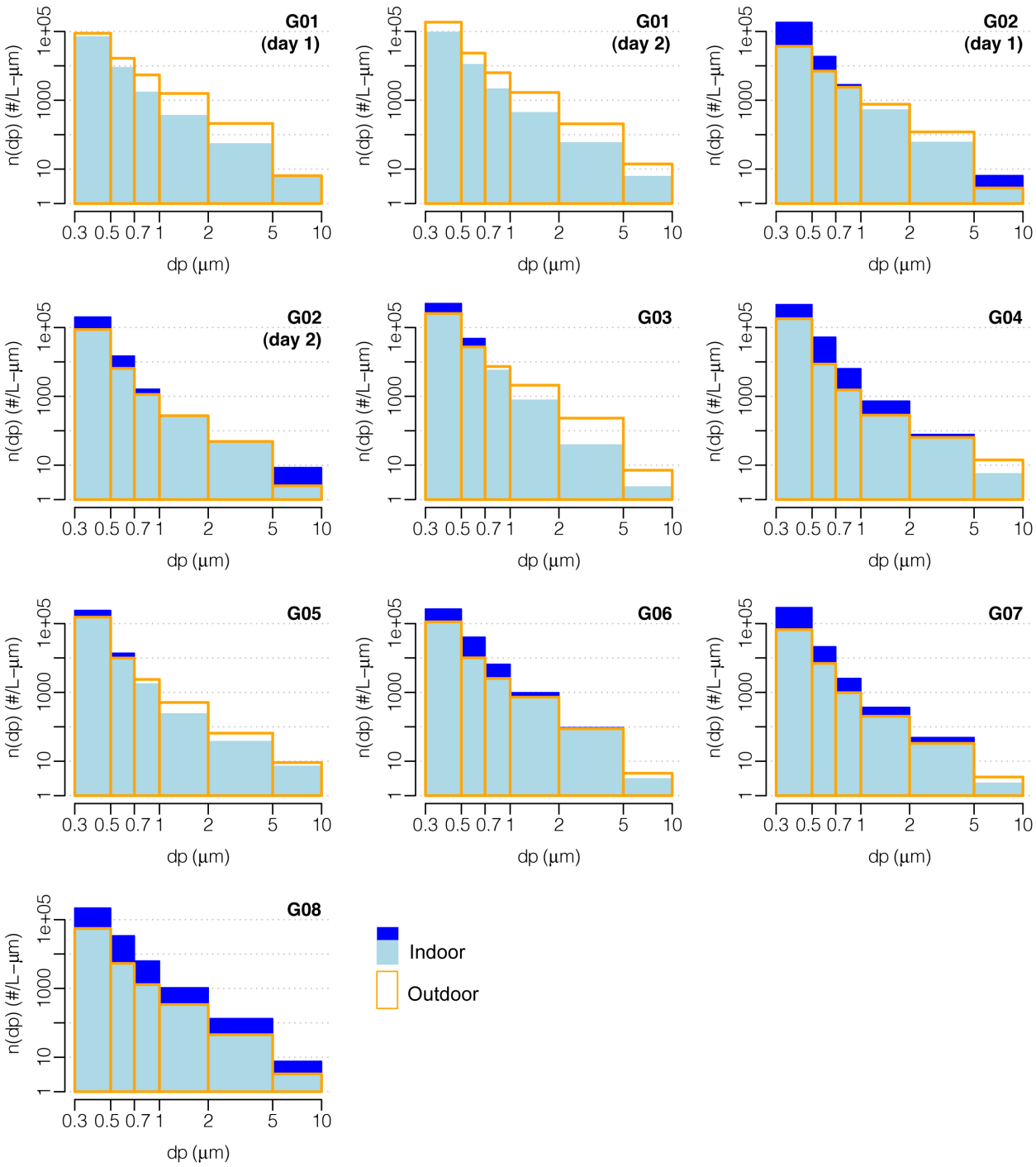

Figure 9 Number distributions of particles between 0.3 and 10 µm measured in eight grocery stores. Darker blue shows the portion of indoor particles that exceeded the outdoor levels (note: both axes shown in log-scale).

In all stores, particles in the smallest size bin, i.e. 0.3 to 0.5 µm had the highest count concentrations. In most of the grocery stores and a few stores of the other two types, indoor particle counts exceeded the outdoor levels, as highlighted in darker blue in the figures. In grocery stores (Figure 9), indoor particle counts exceeded the outdoor levels more notably

among the smaller size bins. This was likely because of fine particles generated from cooking, which occurred throughout the day to prepare ready-to-eat food. All the grocery stores sampled also included a small café area. Cooking activities observed on the sales floor of the grocery stores included stir-frying, grilling, baking, and coffee roasting. In addition, some of the grocery stores also had a separate kitchen area where more cooking likely took place.

In furniture/hardware and apparel stores, indoor particle counts rarely exceeded the outdoors, as shown Figure 10 and Figure 11. In the limited cases where the indoor particle counts exceeded the outdoors, such as in stores F7 and A5, they only occurred in the larger particle size bins.

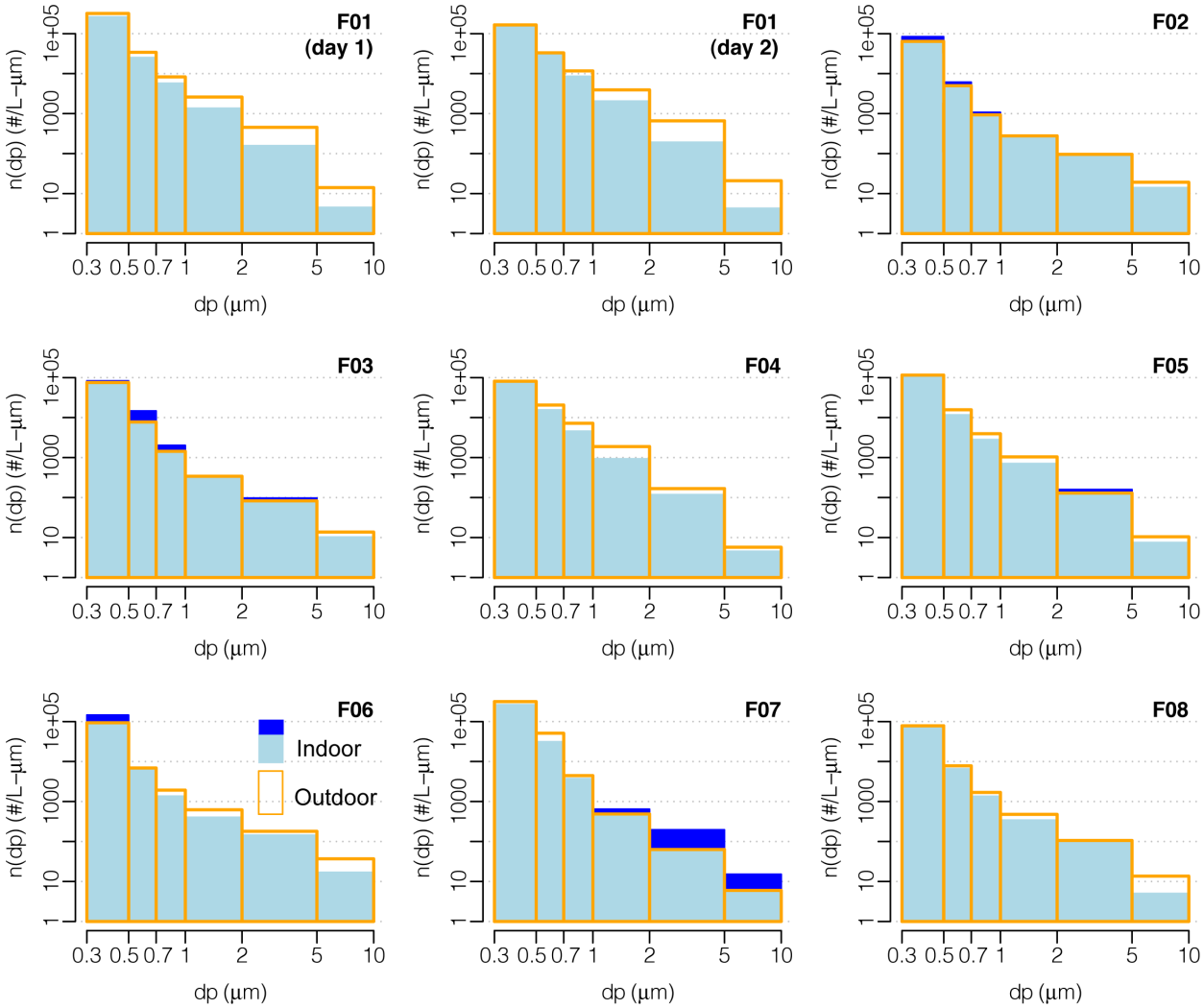

Figure 10 Number distributions of particles between 0.3 and 10 µm measured in eight furniture/ hardware stores. Darker blue shows the portion of indoor particles that exceeded the outdoor levels.

The use of high efficiency air filters can significantly lower the indoor particle levels. G1 is the only grocery store with indoor particle counts substantially lower than the outdoor levels. The store used MERV 15 bag filters, which remove particles much more effectively than the MERV 2 or MERV 8 used in other grocery stores. Better capturing of the cooking emissions by kitchen

range hoods may also explain the lower indoor particle levels measured in G1, but this study did not measure the range hood exhaust flow to determine if their performance is a contributing factor.

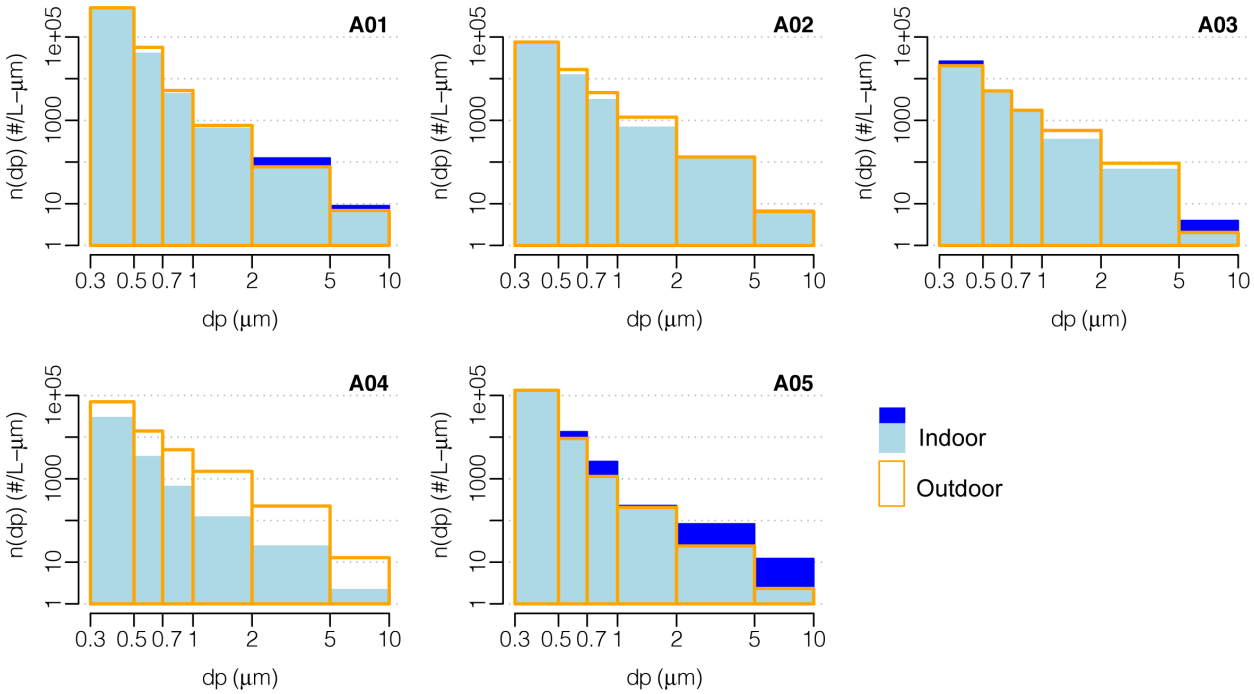

Figure 11 Number distributions of particles between 0.3 and 10 µm measured in five apparel stores. Darker blue shows the portion of indoor particles that exceeded the outdoor levels.

The average indoor and outdoor ultrafine particle counts measured between 10 am and 5 pm are shown in Figure 12. The sampling locations and durations were largely the same as the fine particle counts measured by the optical particle counters. The ultrafine particle counts were collected using a pair of identical water-based condensation particle counters (WCPCs) measuring particles between 6 nm and 3 µm. Even though the WCPCs measured particles in size ranges that overlap with the optical particle counters (0.3 to 10 µm), the number counts measured by the WCPCs were dominated by the smaller particles with diameter between 0.01 and 0.1 µm.

Figure 12 clearly shows that only grocery stores have significant sources of ultrafine particles, likely from cooking. The peak 1-hour indoor counts were typically measured between 12:30 to 1:30 pm. This timing correlated with an increase in cooking activities to serve customers during lunchtime. In furniture/hardware and apparel stores, indoor ultrafine particle counts were generally lower than the outdoor counts. The study of small- and medium-sized commercial buildings in California by Wu et al. (2012) also observed elevated ultrafine particles in two grocery stores that were included in their study.

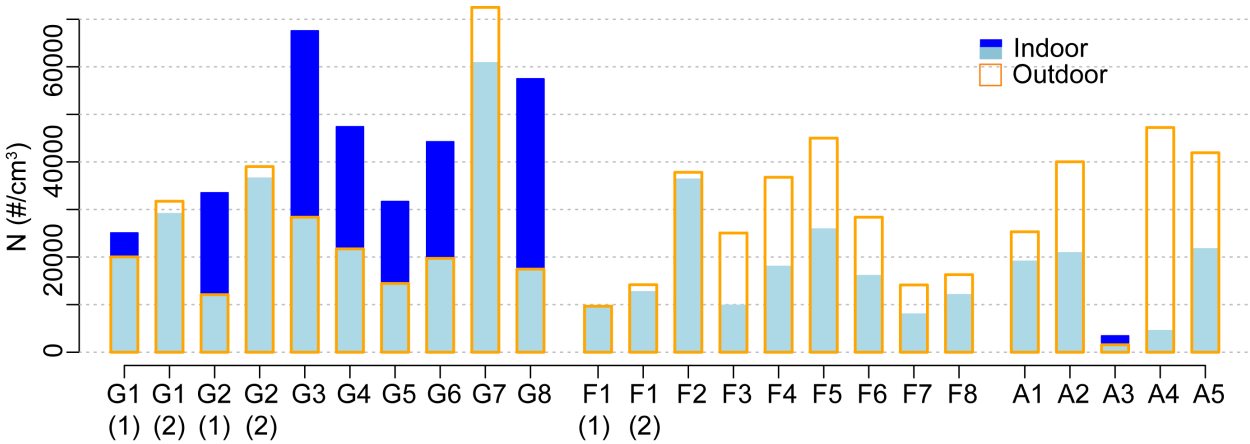

Figure 12 Average ultrafine particle counts measured in grocery (G), furniture/hardware (F), and apparel (A) stores. Darker blue shows the portion of indoor particles that exceeded the outdoor levels. Stores G1, G2, and F1 were sampled on two days: (1) and (2).

Mass concentrations of particles with diameter less than 2.5 μm (PM2.5) and less than 10 μm (PM10) were also measured using a filter-based method during the same sampling period as the real-time particle counters. The average air sample volume collected for measuring the mass concentrations of PM2.5 and PM10 was about 3.9 m^3 (standard deviation = 0.5 m^3). One sample was lost (indoor PM2.5 at store A5) due to instrument error. Table 6 shows the mass concentrations measured from the 21 store visits.

Table 6 shows that a substantial mass fraction of the particles measured indoors was less than 2.5 μm. Using the ratio of PM2.5-to-PM10 as the metric, as shown in the two rightmost columns in Table 6, outdoor particles show an average ratio of 0.45, 0.44, and 0.48 in each of the three types of stores. However, the indoor PM2.5-to-PM10 ratio differed by store types. The indoor PM2.5-to-PM10 ratio has an average of 0.72 in grocery stores, compared to 0.45 in both furniture/hardware stores and apparel stores.

The indoor and outdoor PM2.5 and PM10 mass measurements were plotted in Figure 13. The figure shows that grocery stores generally had indoor particle counts higher than outdoors, as indicated by the blue columns showing indoor PM mass exceeding the height of the corresponding outdoor values (in orange). Most of the particle count data in Figure 9 also show indoor measurements (in blue) exceeding the outdoor values (in orange). Both the particle count data (Figure 9) and the particle mass data (Figure 13) show that G1 had lower indoor PM than outdoors. This is likely because G1 used MERV 15 air filters, which were far more effective at removing PM than the MERV 8 air filters used in the other grocery stores. Besides G1, store G5 also had indoor particle counts similar to the outdoor air (Figure 9), which is again reflected in the mass concentration measurements in Figure 13. Furthermore, Figure 13 also shows that a few of the furniture/hardware stores (F7) and apparel stores (A1 and A5) had higher indoor PM10 than outdoors. The number concentration distributions shown in Figure 10 and Figure 11 also suggest this to be the case.

Table 6 Mass concentrations of PM2.5 and PM10 measured from 21 retail stores visits.

	Sampling Time		PM2.5 ($\mu g/m^3$)		PM10 ($\mu g/m^3$)		PM2.5-to-PM10 Ratio	
	Start	End	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor
Grocery Stores								
G1 (1) *	10:30	16:55	11.1	8.0	21.3	10.5	0.52	0.76
G1 (2) *	8:45	16:05	20.7	6.6	40.0	12.1	0.52	0.54
G2 (1) *	9:35	15:00	4.5	10.8	24.7	18.4	0.18	0.59
G2 (2) *	8:35	16:15	4.3	12.3	8.1	18.7	0.54	0.66
G3	9:40	17:15	16.8	32.2	29.7	46.7	0.56	0.69
G4	10:55	17:30	9.6	17.6	23.6	19.7	0.41	0.90
G5	10:50	17:15	9.6	9.2	18.5	13.4	0.52	0.69
G6	10:45	17:00	5.6	16.5	11.4	20.6	0.49	0.80
G7	11:00	17:25	2.6	19.3	7.7	22.6	0.34	0.85
G8	11:05	17:20	0.5	14.0	9.4	28.9	0.06	0.48
Furniture / Hardware Stores								
F1 (1) *	12:20	17:30	12.5	11.0	24.2	17.3	0.52	0.64
F1 (2) *	11:40	16:40	14.2	13.9	32.4	21.3	0.44	0.65
F2	10:55	17:10	6.8	6.1	18.5	19.4	0.37	0.31
F3	10:35	17:05	5.9	6.0	21.1	16.1	0.28	0.38
F4	10:40	17:50	14.8	6.9	20.3	12.5	0.73	0.55
F5	9:40	16:40	12.5	8.0	23.6	20.3	0.53	0.39
F6	11:05	17:05	13.8	8.3	48.3	20.8	0.28	0.40
F7	10:45	17:10	8.0	12.5	18.1	26.7	0.44	0.47
F8	10:45	17:00	9.6	3.2	26.0	12.6	0.37	0.26
Apparel Stores								
A1	10:50	16:35	13.7	14.1	24.7	29.9	0.55	0.47
A2	12:25	17:40	7.6	7.0	18.4	17.2	0.41	0.41
A3	10:05	17:35	3.8	3.9	10.7	11.4	0.35	0.34
A4	12:00	16:50	16.3	4.2	37.5	7.5	0.43	0.57
A5	11:55	18:05	6.4	--	10.1	11.5	0.39	--

* Stores G1, G2, and F1 were measured on two days, as indicated by (1) and (2).

Because particle mass were only sampled for a short duration of about 6 hours, the values shown in Table 6 may not be representative of the daily or longer time average mass concentrations. Even so, it is useful to compare the particle mass measurements with the 24-hour and annual PM2.5 and PM10 standards to evaluate if the levels found in retail stores were of potential health concerns.

In California, the Air Resources Board adopted the U.S. EPA 24-hour PM2.5 national ambient air quality standard of 35 $\mu g/m^3$ for the 98th percentile of measured values. The PM2.5 mass measurements were less than 35 $\mu g/m^3$ in all the sampled retail stores. The California 24-hour PM10 ambient air quality standard is 50 $\mu g/m^3$. Again, all the sampled stores met this standard. Based on these comparisons, the short-term health risks of exposure to PM2.5 and PM10 in retail stores may be low; however, little is known about the potency of indoor-generated particles (other than allergens and those linked to infections) to cause adverse health effects. In stores G1, G2, and F1 where sampling lasted for two days and the particle counts were monitored continuously overnight, lower particle counts were measured when the stores were closed. As a result, the 24-hour average particle mass concentrations would likely be lower than the values shown in Table 6.

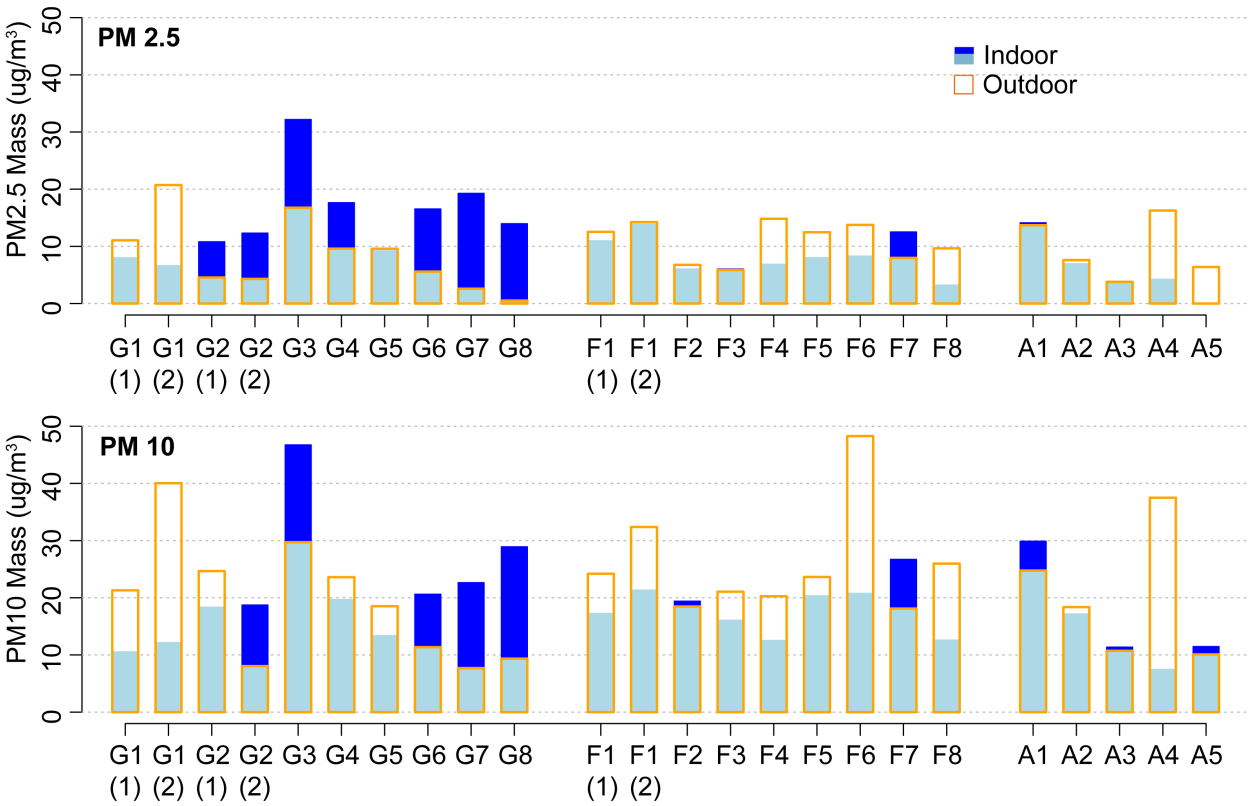

Figure 13 Mass concentrations of PM2.5 and PM10 measured by a filter-based gravimetric method. Darker blue shows the portion of indoor particles that exceeded the outdoor levels. Stores G1, G2, and F1 were sampled on two days: (1) and (2).

The California annual PM2.5 ambient air quality standard is 12 μg/m^3. For PM10, the annual standard is 20 μg/m^3. These ambient air quality standards were implemented in 2002 in California. In December 2012, U.S. EPA lowered the PM2.5 annual standard from 15 μg/m^3 to 12 μg/m^3, but the PM10 annual standard remained at 50 μg/m^3. There are reasons to expect that the PM2.5 and PM10 mass concentrations shown in Table 6 may not represent the long-term averages needed for comparison with the annual standards. It is because there are daily, weekly, and seasonal variations in retail store operation and occupancy that are not reflected in this data. Nonetheless, the standards of 12 μg/m^3 for PM2.5 and 20 μg/m^3 for PM10 can be viewed as health guidelines that protect the most sensitive individuals from long-term exposure risks. From this viewpoint, if indoor-generated PM2.5 poses a similar level of health risk as outdoor air PM2.5, indoor PM2.5 may pose some long-time health risks in half of the grocery stores that were sampled. In furniture/hardware and apparel stores, the indoor PM2.5 levels were generally lower indoors than outdoors. This means that the long-term exposure risks to PM2.5 inside those stores are likely of lesser concerns relative to the outdoors. The long-term exposure risks to PM10 are comparable or lower indoors relative to the outdoors in all store types.

Optical particle counters are much lower in costs than other continuous instruments, such as tapered element oscillating microbalance (TEOM) and beta attenuation monitor (BAM). Optical particle counters are also substantially cheaper to operate than filter-based methods, because

weighing of filters is very labor intensive. However, mass concentrations are necessary for exposure assessment because health guidelines and past literature are largely based on that metric. There are many assumptions to be made to estimate particle mass concentrations from count measurements, such as effective diameter of the particles in a given size bins and particle density. An analysis is performed here in an attempt to estimate PM2.5 and PM10 mass concentrations from the real-time data collected by both the optical particle counters and the WCPCs.

In this study, all three types of measurements were collected concurrently with slight differences in the start and end time of no more than half hour of one another. Each set of the three outdoor and three indoor instruments were also co-located. Even though they did not share a common sampling port, this is unlikely to be a key source of measurement error when data were averaged over a few hours.

Particle mass concentrations, M (μg/m^3), were estimated from the particle count measurements, N (#/m^3), as follows:

$$M = \sum_i \frac{\pi}{6} \rho \left(d_i^* \right)^3 N_i$$

Eq 5

where $\rho = 1.65$ μg/cm^3 is the particle density, d_i^* (μm) is the equivalent particle diameter for each size bin i, and N_i is the particle count concentration.

The equivalent particle diameter d_i^* gives the same total volume of particles assuming that the number of particles are uniformly distributed within the size bin i that is bounded by the lower and upper diameters, $d_{i,a}$ and $d_{i,b}$ (μm).

$$\left(d_i^* \right)^3 N_i = \int_{d_{i,a}}^{d_{i,b}} \frac{N_i}{\left(d_{i,b} - d_{i,a} \right)} x^3 \, dx$$

$$d_i^* = \left[\frac{d_{i,b}^4 - d_{i,a}^4}{4\left(d_{i,b} - d_{i,a} \right)} \right]^{1/3}$$

Eq 6

where x is a variable representing the diameters of particles that are uniformly distributed between $d_{i,a}$ and $d_{i,b}$ (μm).

The mass of PM2.5 is estimated from the four smallest size bins measured by the optical particle counters: 0.3–0.5 μm, 0.5–0.7 μm, 0.7–1 μm, and 1–2 μm. Within each size bin, d_i^* is calculated according to Eq 6. The resulting values of d_i^* for the four size bins are 0.41, 0.61, 0.86, and 1.6 μm.

PM10 is estimated from the sum of all six size bins measured by the optical particle counters, i.e. the four size bins listed above for PM2.5 plus two more: 2–5 μm and >5 μm. It is assumed that

there were very few particles larger than 10 μm that were counted by the optical particle counters. Using Eq 6, the resulting values of d_i^* for the two largest size bins are 3.7, and 7.8 μm, respectively.

For the portion of particles in the ultrafine range, it is not possible to estimate an effective diameter using Eq 6 because the water-based condensation particle counters (WCPCs) only gave the total counts of particles between 6 nm and 3 μm. To estimate an effective diameter d_e (μm) of ultrafine particles needed for calculating their mass, a regression was performed as described in Eq 7.

$$M_{PM2.5} - M_{0.3-2\mu m} = N_{UFP} \frac{\pi}{6} \rho d_e^3$$

Eq 7

where $M_{PM2.5}$ is the measured PM2.5 mass concentrations, $M_{0.3\text{-}2um}$ is the estimated mass of particles between 0.3 and 2 μm, and N_{UFP} (#/cm^3) is the measured ultrafine particle counts by WCPC. This approximation assumes that the difference between $M_{PM2.5}$ and $M_{0.3\text{-}2um}$ is all due to the mass of ultrafine particles with diameters less than 0.3 μm. In addition, Eq 7 assumes that this mass of particles less than 0.3 μm can be reasonably predicted using the ultrafine particle counts measured by the WCPC.

Figure 14 shows a roughly linear relationship between N_{UFP} and the difference between $M_{PM2.5}$ and $M_{0.3\text{-}2um}$, as described in Eq 7. Linear regression gives a slope of 2.54×10^{-4} and $R^2 = 0.75$. This simple relationship appears to be applicable for both outdoor and indoor particles measured in all retail stores sampled from 21 store visits. Assuming that ultrafine particles have the same particle density of 1.65 g/cm^3 as the larger particles, the best-fitted slope from the linear regression gives d_e = 0.066 μm.

Using 0.066 μm as the equivalent diameter for the ultrafine particles, the mass concentrations of ultrafine particles are estimated and then added to the optical particle counter results. Figure 15 compares the predicted PM2.5 and PM10 mass concentrations from the sum of the ultrafine particles and the applicable size bins of the optical particle counters (x-axis), with measured values from the filter-based method (y-axis). The predicted indoor PM2.5 mass concentrations agreed well with measurements, as shown in Figure 15(a). The blue dotted line has a slope of 1.02 and $R^2 = 0.70$; see Table 7 for the regression results describing the agreement between the predicted and measured PM2.5 and PM10 mass.

However, the predictions of outdoor PM2.5 mass do not agree well with measurements, as shown in Figure 15(a). This may be because outdoor fine particles contained a higher fraction of volatile compounds than indoor particles, which were lost from the filter-based measurements. Assumptions about particle density and their equivalent diameters also contributed to the uncertainty in the particle mass predictions.

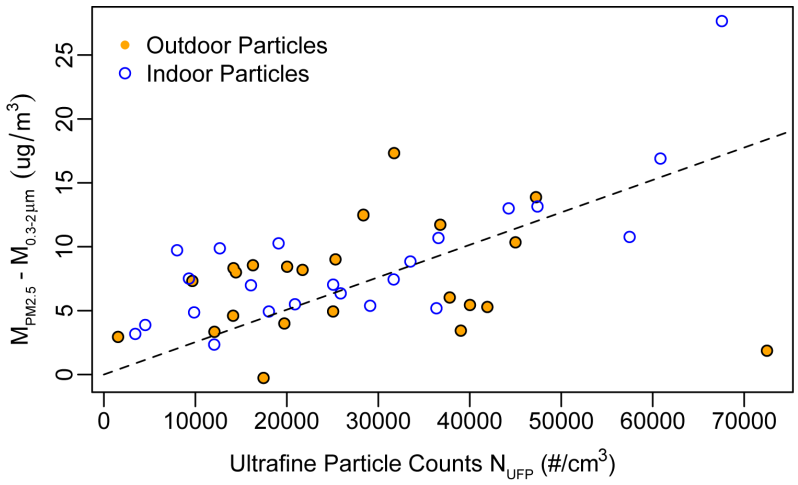

Figure 14 Relationship between the ultrafine particle counts measured by WCPCs (x-axis) and the predicted mass of particles <0.3 µm (y-axis). A total of 48 data points are shown here, including 21 store visits, and three stores (G1, G2, and F1) sampled for two days.

The predictions of outdoor and indoor PM10 mass generally agreed with measurements, as shown in Figure 15(b) by the regression slope near unity. But, there are considerable scattering between the measured and predicted values, which is reflected in the relatively low R^2 values of about 0.5 (see Table 7).

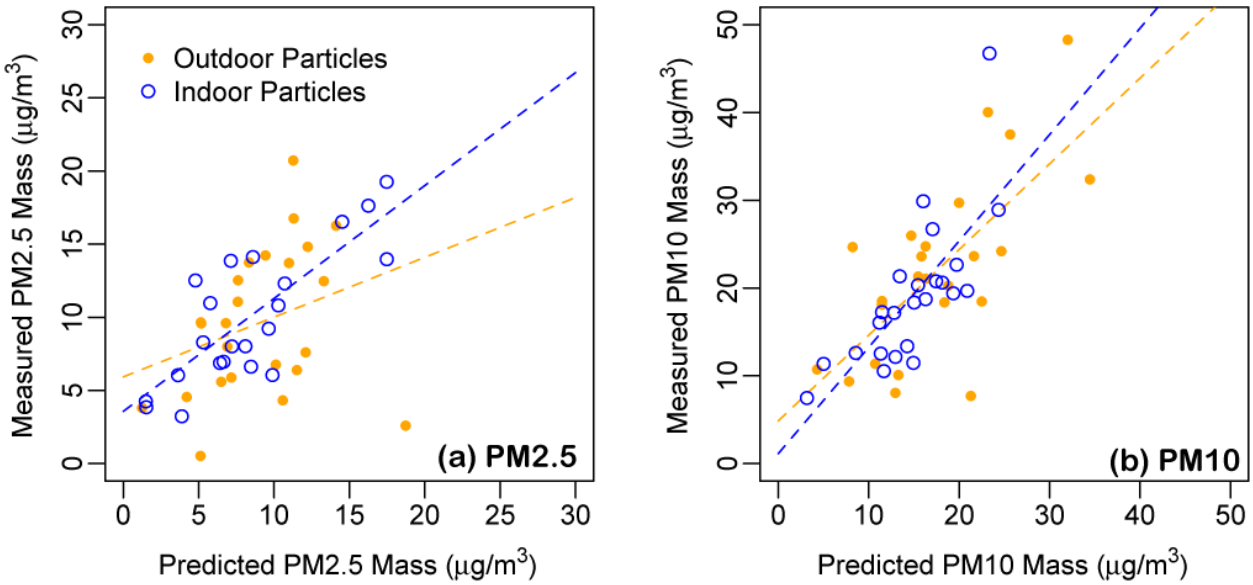

Figure 15 Comparison of the measured and predicted (a) PM2.5 and (b) PM10 mass concentrations. Particle mass was measured using filter-based method analyzed gravimetrically. Predictions of the mass concentrations were by summing the contributions from the optical particle counters and WCPCs.

Overall, the predictions from particle count measurements give reasonable estimates of particle mass, especially in terms of summary statistics, such as the mean PM2.5 and PM10 mass concentrations in retail stores as shown in Table 7. However, the estimates are less reliable on a per-store basis.

Table 7 Comparison of the measured and predicted PM2.5 and PM10 mass concentrations.

		Mass Concentration (μg/m^3) Mean (Standard Deviation)		Linear Regression Results		
		Measured	**Predicted**	**Slope**	**Intercept**	**R^2**
PM2.5	Outdoor	9.6 (5.1)	9.1 (3.8)	0.41	5.93	0.053
	Indoor	10.9 (6.4)	8.9 (5.2)	1.02	1.73	0.70
PM10	Outdoor	22.0 (10.3)	17.6 (7.3)	0.98	4.86	0.46
	Indoor	19.0 (8.3)	14.6 (5.1)	1.21	1.12	0.54

Using the method described above, the mass fraction of particles <0.3 μm, 0.3–2 μm, and 2–10 μm can be estimated from the count measurements collected using the optical particle counters and WCPCs, as shown in Figure 16.

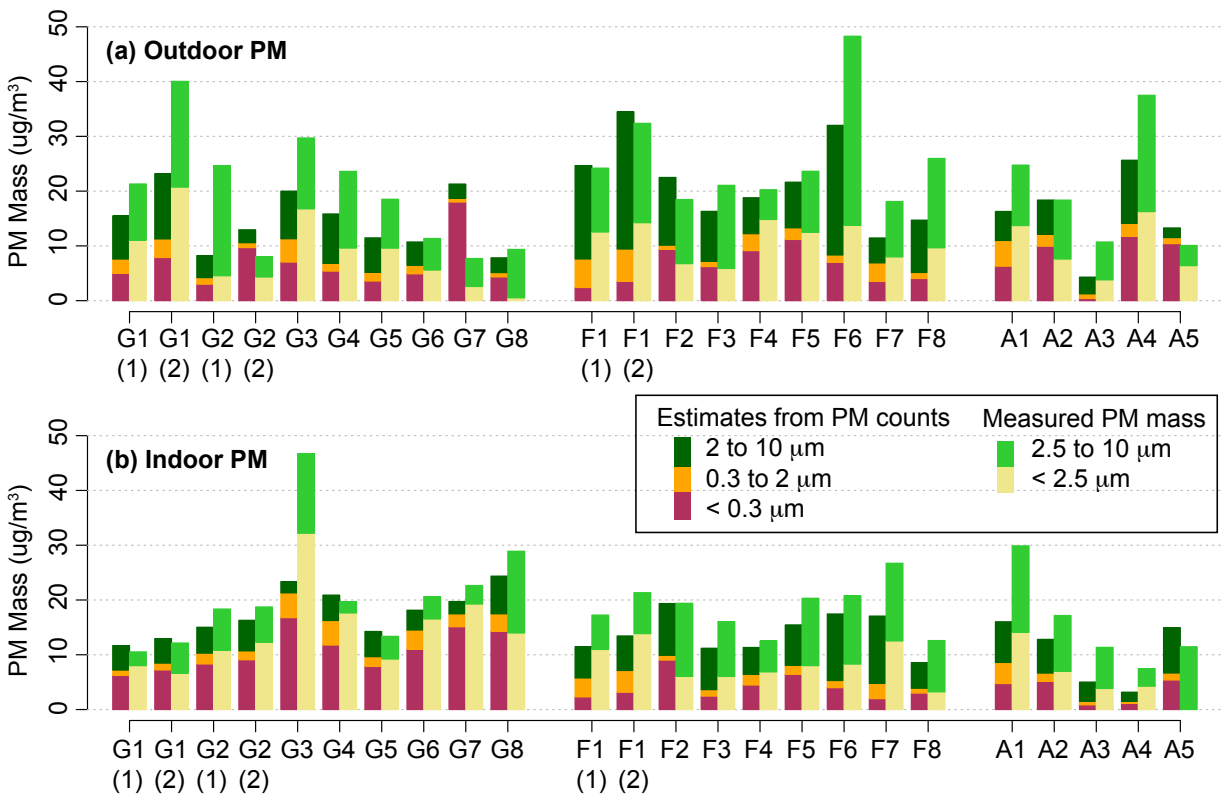

Figure 16 Comparison of the measured and estimated mass concentrations of PM in different particle size ranges. Stores G1, G2, and F1 were sampled on two days: (1) and (2).

Figure 16 shows that a substantial fraction of the particle mass were found in the small size range of < 0.3 μm, especially inside grocery stores. On average, 60% of the PM mass in grocery stores

is estimated to be from particles < 0.3 μm. In stores without significant particle sources (i.e., furniture/hardware and apparel stores), only 30% of the PM mass is estimated to be from particles < 0.3 μm. The higher mass fractions of PM < 0.3 μm estimated in grocery stores suggest that particles emitted from cooking in grocery stores tend to be small, with diameters in the 0.1 μm size range and less. This observation agrees with the particle mass distributions that were reported in literature from studies characterizing particle emissions from cooking (Torkmahalleh et al., 2012).

In urban areas, outdoor particles in the accumulation mode (0.1 to 1 μm) can make up a large fraction of the particle mass fraction. The measured mass of PM2.5 and PM10 in Figure 16 show that there were in most cases roughly equal proportions of outdoor particle mass in the <2.5 μm and the 2.5 to 10 μm bins. Since there was no major source of indoor PM in furniture/hardware and apparel stores, a significant portion of the indoor PM likely originated from outdoors. As a result, roughly the same portions of PM mass fraction were found in these two store types as in outdoor air.

5.6 Volatile Organic Compounds

Table 8 shows a list of 48 VOCs that were sampled in the retail stores. Acrolein was only sampled in a subset of stores as a preliminary effort to see how well the pentafluorophenyl hydrazine (PFPH) method can be adopted to field sampling. Results of acrolein concentrations sampled from five grocery stores are presented last. Concentrations of other VOCs listed in Table 8 were measured in each of the 21 store visits.

Table 8 List of volatile organic compounds sampled in 21 retail stores.

Alcohols 2-Ethylhexanol 2-Butoxyethanol	**Aldehydes** Formaldehyde Acetaldehyde Acrolein Hexanal Octanal Decanal Butanal Heptanal Nonanal Benzaldehyde	**Aromatics** Benzene Toluene Ethylbenzene o-Xylene Styrene Butylbenzene Naphthalene m/p-Xylene Phenol	**Esters** Texanol* TXIB** Diethyl phthalate Dibutyl phthalate Dimethyl phthalate Cyclotrisiloxane (D3-Siloxane) Cyclotetrasiloxane (D4-Siloxane) Cyclopentasiloxane (D5-Siloxane)
Alkanes Heptane Octane Decane Undecane Dodecane Tetradecane Hexadecane		**Halogenated alkanes** Tetrachloroethylene Trichloromethane	**Halogenated aromatics** 1,2,4-Trimethylbenzene 1,2,3-Trimethylbenzene 1,4-Dichlorobenzene
Ketone Acetone	**Terpenoids** d-Limonene a-Pinene a-Terpineol 3-Carene g-Terpinene	**Misc** 1-Methyl-2-pyrrolidinone	

* Texanol is 2,2,4-Trimethyl-1,3-pentanediol monoisobutyrate.
** TXIB is 2,2,4-Trimethyl-1,3-pentanediol diisobutyrate.

A duplicate sample was always collected. Comparison of the duplicate samples shows that on average, they agreed to within 15% of each other. The average difference in concentration of the duplicates divided by their mean was 13.5% for the outdoor samples, and 15.2% for the indoor samples. Samples with concentrations below the minimum detection limit (MDL) were not included in the comparison of duplicates.

The Carbopack sorbent tubes used typically had a MDL of about 0.3 $\mu g/m^3$ for a 6-m^3 air sample, with the range of MDL between 0.04 $\mu g/m^3$ (1,4-dichloro-benzene) and 1.1 $\mu g/m^3$ (D3-siloxane), as shown in Table 9. DNPH analysis by HPLC for formaldehyde, acetaldehyde, and acetone has very low MDL of <1 ng/m^3.

Table 9 Minimum detection limits of VOC samples.

Carbopack Sorbent Tube MDL ($\mu g/m^3$)				
1,4-Dichlorobenzene	0.04		Trichloromethane	0.19
1,2,4-Trimethylbenzene	0.04		1-Methyl-2-pyrrolidinone	0.20
Naphthalene	0.05		Tetradecane	0.20
1,2,3-Trimethylbenzene	0.05		Heptane	0.21
3-Carene	0.05		2-Ethylhexanol	0.21
Decane	0.05		Hexadecane	0.25
Butylbenzene	0.06		Octane	0.25
d-Limonene	0.07		Tetrachloroethylene	0.32
a-Pinene	0.08		Benzene	0.33
Styrene	0.09		Dibutyl phthalate	0.34
Undecane	0.11		Toluene	0.35
Benzaldehyde	0.11		Texanol	0.37
D5-Siloxane	0.12		Dimethyl phthalate	0.37
Octanal	0.12		Ethylbenzene	0.39
Decanal	0.12		o-Xylene	0.40
Heptanal	0.13		m/p-Xylene	0.44
a-Terpineol	0.13		Diethyl phthalate	0.51
g-Terpinene	0.15		Hexanal	0.53
Nonanal	0.15		Butanal	0.55
2-Butoxyethanol	0.17		Phenol	0.56
Dodecane	0.19		TXIB	0.69
D4-Siloxane	0.19		D3-Siloxane	1.11
DNPH Aldehyde Analysis MDL (ng/m^3)				
Formaldehyde	0.03		Acetone	0.28
Acetaldehyde	0.06			

In each of the stores, indoor concentrations were measured in up to four locations, depending on the store size, as shown in Table 10. VOCs samples were collected in the afternoon, typically between 1 pm and 3 pm. The sampling times shown in Table 10 reflect the start of the first VOCs sample collected from the retail store, and the end time of the last sample. In some of the stores, sorbent tubes and aldehyde cartridge samples were collected sequentially, thus explains the longer time difference between the start and end time reported in Table 10.

Table 10 VOCs sampling times and number of indoor sampling locations.

	Sampling Time Outdoor		Sampling Time Indoor		# Indoor Sample Location		Sampling Time Outdoor		Sampling Time Indoor		# Indoor Sample Location
	Start	End	Start	End			Start	End	Start	End	
Grocery Stores						**Furniture / Hardware Stores**					
G1 (1)*	14:20	16:55	14:05	16:55	4	F1 (1)*	16:25	17:20	14:05	16:25	3
G1 (2)*	13:05	15:30	12:45	15:15	4	F1 (2)*	12:40	13:40	12:30	15:10	3
G2 (1)*	12:40	14:45	12:20	15:05	4	F2	15:10	16:10	12:50	15:10	3
G2 (2)*	12:25	14:40	12:15	14:55	4	F3	12:35	15:40	12:25	15:35	3
G3	12:45	14:10	12:10	13:55	4	F4	13:35	14:55	13:15	14:45	4
G4	13:00	16:25	12:25	16:15	4	F5	12:15	13:55	12:05	13:55	2
G5	12:50	16:50	14:35	16:25	4	F6	15:20	16:30	14:45	16:05	4
G6	12:15	13:35	12:05	13:30	4	F7	14:05	15:35	13:45	15:30	4
G7	12:20	13:45	12:00	13:40	4	F8	13:00	14:30	12:55	14:30	4
G8	12:40	14:10	12:20	14:05	3						
Apparel Stores											
A1	13:05	14:20	13:15	14:15	1						
A2	13:40	14:40	13:35	15:35	1						
A3	12:10	13:15	12:00	14:15	3						
A4	15:20	16:55	14:55	16:45	3						
A5	14:10	15:40	14:05	15:35	2						

* Stores G1, G2, and F1 were measured on two days, as indicated by (1) and (2).

Concentration distributions of indoor VOCs are shown in Figure 17. Each boxplot includes up to 79 pairs of duplicated measurements (see Table 10 for the number of indoor sampling locations). VOCs are ordered by the median indoor concentrations measured, with acetone being the highest on the top left, and naphthalene being the lowest on the bottom right. VOCs plotted on the right-frame of Figure 17 were low in indoor concentrations typically less than 1 $\mu g/m^3$. On the other hand, VOCs plotted on the left-frame of Figure 17 exceeded 1 $\mu g/m^3$ indoors more often than not.

The red circles indicate the mean outdoor concentrations for VOCs above MDL. Only about 10 VOCs were routinely above MDL measured outdoors. They include acetone, acetaldehyde, formaldehyde, toluene, xylene, benzene, heptane, decane, 1,2,4-trimethylbenzene, and benezaldehdye. These chemicals were all associated with combustion, most likely from vehicle emissions.

The outdoor concentrations shown in red circles for heptane and benzene overlapped with the indoor concentrations represented by the boxplot, meaning that the outdoor and indoor concentrations tended to be similar for these two compounds. But, chemicals such as formaldehyde, acetaldehyde, and benzaldehdye, often had indoor concentrations that were substantially higher than the outdoors. This comparison of the outdoor and indoor VOC measurements indicates that these compounds had indoor sources.

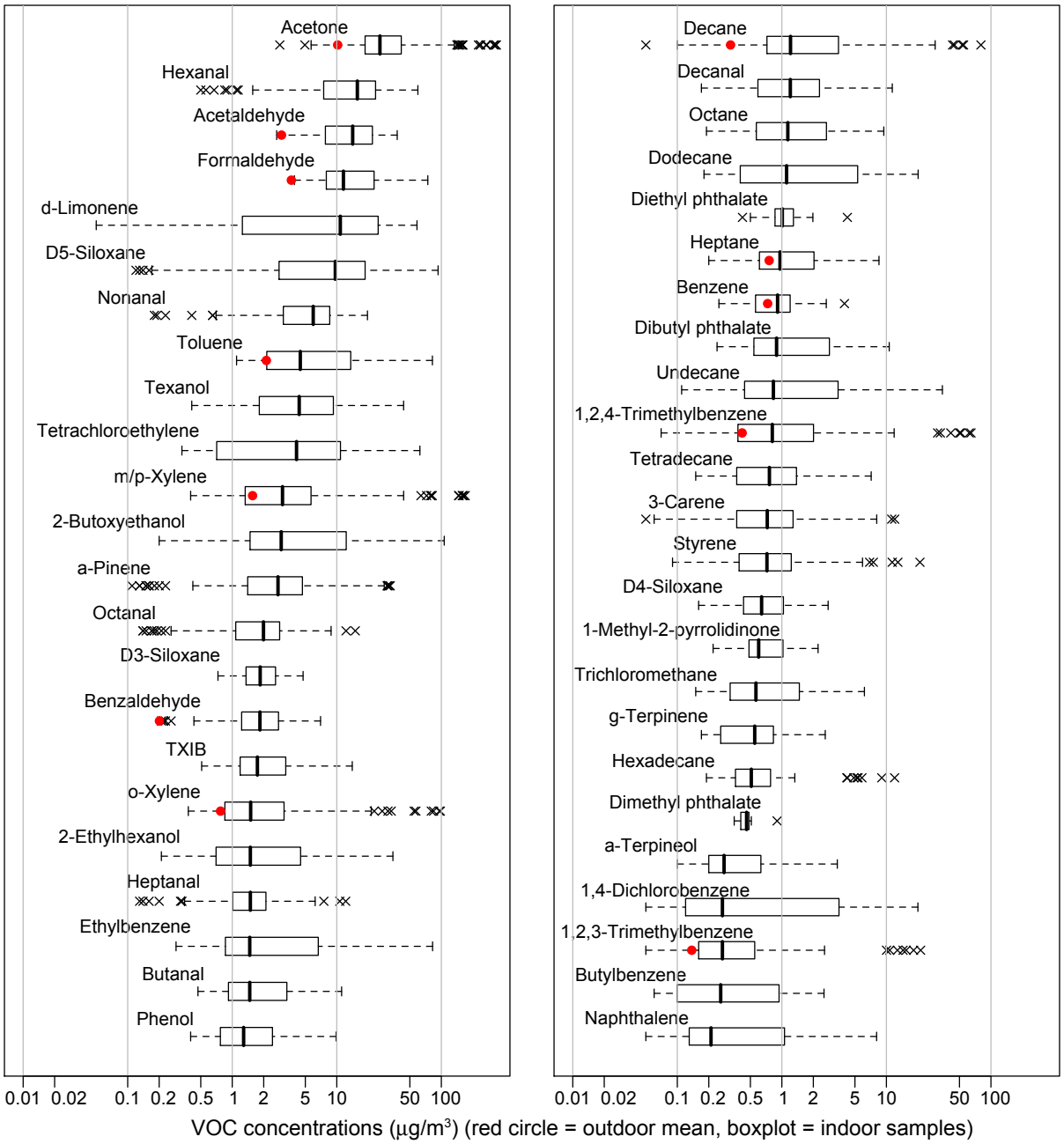

Figure 17 Summary of the outdoor (red circle) and indoor VOC concentrations (boxplot) measured from 21 retail store visits. VOCs are plotted in the order of the median indoor concentrations measured. Concentrations outside the range of typical values are indicated by the "x" symbols.

In stores where VOCs were measured at multiple locations, the data shows large spatial differences in the concentrations measured. Figure 18 shows the difference between the maximum and minimum concentrations measured as a percentage of the mean concentration found in the store. Four smaller stores, F5, A1, A2, and A5 were excluded from this analysis

because VOCs were measured at one or two locations only. This leaves data from 17 stores shown in Figure 18, among them three of the stores (G1, G2, and F1) were sampled on two days. Only VOCs that were frequently measured above MDLs were plotted in Figure 18.

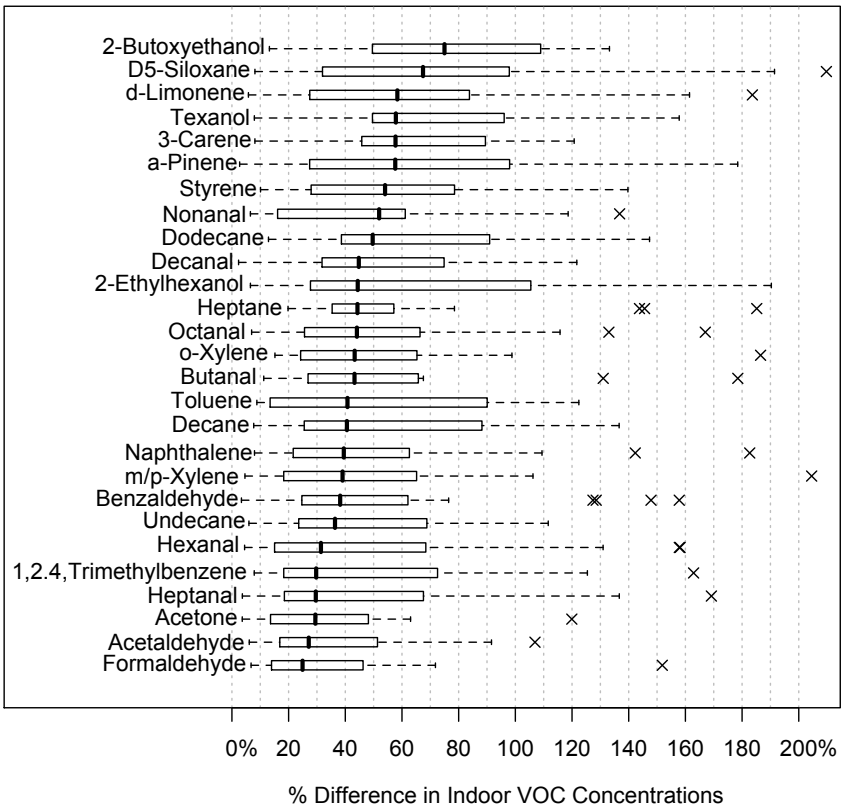

Figure 18 Spatial differences in VOC concentrations plotted as the difference in maximum and minimum concentrations as a percentage of the mean. This plot only includes stores where VOCs were measured at three or more locations, and compounds that were frequently measured above MDLs. Values outside of the typical range are indicated by the "x" symbols.

Figure 18 shows that formaldehyde, which is largely emitted from building materials, has the lowest spatial differences among all VOCs. But even for formaldehyde, the spatial difference in indoor concentrations was significant. The percent differences from spatial variability typically range between 15% and 45%, with a median difference of 25%. Compounds with significant outdoor sources, such as toluene and xylene, showed higher spatial variability than formaldehyde, with a median value of about 40%. Compounds that are associated with specific product use or store merchandise, such as a-pinene, d-limonene, and D5-siloxane, showed the highest spatial differences. For these compounds, different siting of the sampling instruments could result in 60% differences in the measured indoor concentrations.

Another way to identify VOCs of potential interests is by comparing the measured concentrations with health guidelines and odor thresholds. Table 11 shows a list of VOCs with level of concerns that may be reached in some retail stores. It also includes VOCs that have low

odor thresholds, such as hexanal and octanal. Of the VOCs listed in Table 11, the lowest concentration at which exposure to a compound may raise health concern is acrolein. Since acrolein is an important contaminant of concern and the sampling method used to measure its concentrations was newly developed for this work, more detail discussions of this VOC are presented later in Section 5.7 of this report.

Table 11 Chronic health guidelines and odor thresholds of VOCs with indoor concentration that may reach level of concerns.

	Chronic Health Guideline (μg/m^3)		**Odor Thresholds (μg/m^3)** *
	Most stringent	**Other guideline**	
Acrolein	0.02 μg/m^3 (EPA)	0.09 μg/m^3 (ATSDR)	
Naphthalene	3 (EPA)	9 (OEHHA)	80
Acetaldehyde	9 (EPA)	140 (OEHHA)	340
Formaldehyde	9 (OEHHA)	--	90
Benzene	30 (EPA)	60 (OEHHA)	--
Tetrachloroethylene	35 (OEHHA)	--	--
m/p-Xylene	100 (EPA)	700 (OEHHA)	--
Toluene	300 (OEHHA)	5,000 (EPA)	--
o-Xylene	700 (OEHHA)	--	--
Octanal	--	--	2
Hexanal	--	--	32
Benzaldehyde	--	--	180

* Odor thresholds listed here are from Cain and Schmidt (2009), Hodgson and Levin (2003), and Nagata (2003).

Table 12 VOC concentrations measured inside and outside stores (total counts = 24).

	Outdoor Concentration (μg/m^3)		**Indoor Concentration (μg/m^3)**	
	Mean (Min–Max)	**Counts >MDL**	**Mean (Min–Max)**	**Counts >MDL**
Naphthalene	0.1 (0.1–0.3)	4	1.0 (0.1–6.4)	22
Acetaldehyde	3.0 (1.5–8.0)	24	13.6 (4.3–28)	24
Formaldehyde	3.7 (1.4–6.8)	24	18.8 (5.1–59)	24
Benzene	0.7 (0.3–1.5)	15	0.9 (0.3–2.4)	23
Tetrachloroethylene	2.8 (1.1–5.6)	3	5.5 (0.3–22)	10
m/p-Xylene	1.5 (0.5–6.9)	16	14.4 (0.4–160)	24
Toluene	2.1 (0.4–7.9)	21	12.0 (1.4–53)	24
o-Xylene	0.7 (0.4–2.3)	11	7.5 (0.4–90)	21
Octanal	0.3 (0.1–0.7)	5	2.0 (0.2–5.9)	24
Hexanal	0.8 (0.5–1.3)	3	16.1 (0.7–58)	24
Benzaldehyde	0.2 (0.1–0.3)	10	2.0 (0.2–4.8)	24

Table 12 shows the mean and the concentration range of VOCs with potential health or odor concerns measured from 21 store visits. Three stores (G1, G2, and F1) were visited on two sequential days, resulting in a total of 24 sets of store averaged VOC concentrations. Of the eleven VOCs listed in Table 11, the indoor concentrations measured were typically above MDLs with the exception of tetrachloroethylene. But even though many of these VOCs were commonly found indoors, only a few of them were found at levels that may cause health or odor concerns. For example, indoor concentrations of benzene, toluene, o-xylene, and benzaldehyde were all below concentrations that may cause health or odor concerns. Indoor concentrations of naphthalene and m/p-xylene exceeded the most stringent chronic health guideline in one or two

stores only. Similarly, hexanal was measured at a significant level indoor only in one store that had a concentration exceeding the odor threshold. Formaldehyde, acetaldehyde, and octanal are the only VOCs exceeding health guideline or odor threshold in more than a few of the stores that were sampled.

Figure 19 shows the indoor concentrations of six VOCs with at least one store exceeding the most stringent health guideline or odor threshold. Formaldehyde concentrations measured in furniture/hardware stores tended to be higher than in the other two store types. Merchandise containing composite wood products are likely a key indoor source of formaldehyde. But overall, the formaldehyde concentrations found in stores were low compared to levels found in other buildings, possible evident of the effectiveness of emissions standards that were recently implemented in California. For comparison, a median concentration of 36 $\mu g/m^3$ was found in 108 single-family homes built between 2002 and 2004 in California (Offermann, 2009). In January 2009, the first formaldehyde emission standards from composite wood products were implemented in California (Phase I, CARB 2012). Since then, the emission standards have been lowered to more stringent levels (Phase II). Phase I and II compliance labels were found on merchandise in the furniture stores that were sampled. The mean concentration of formaldehyde measured in this study was 19 $\mu g/m^3$.

Apparel store A2 was recently renovated when it was sampled. It had one of the highest formaldehyde concentrations, likely from the building materials used. Yet, there are also other newly opened or recently renovated stores, such as G6, G8, A3, and A5, that had formaldehyde concentrations below the health guideline of 9 $\mu g/m^3$. For example, A3 is a nationwide chain store that considers low emission materials in their choice of interior furnishing and display. Very low formaldehyde concentrations were measured in store A3. Store A4 is from the same chain but located in another part of California. It also had low formaldehyde level (12 $\mu g/m^3$). Most of the grocery stores sampled in this study also had low concentrations of formaldehyde meeting California's stringent health standard. One plausible explanation is because building materials common in grocery stores, such as metal shelves and concrete flooring, do not contain much formaldehyde.

Acetaldehyde concentrations in grocery stores were higher than in other store types likely because of baking (Loh, 2006). Acetaldehyde can also be emitted from building materials. However, Figure 19 shows that the acetaldehyde concentrations in furniture and apparel stores were generally lower than in grocery stores. This suggests that indoor sources associated with grocery stores, such as from baking, likely dominate over sources related to building materials that are more ubiquitous in all buildings.

Octanal and hexanal are examples of two higher molecular weight aldehydes that are odorous at low levels. The very low odor thresholds used in this report are from Cain and Schmidt (2009), Hodgson and Levin (2003), and Nagata (2003). There are significant differences in the threshold values reported in the literature because of variations in experimental procedures and definition of odor thresholds. The threshold values used here are the most sensitive ones.

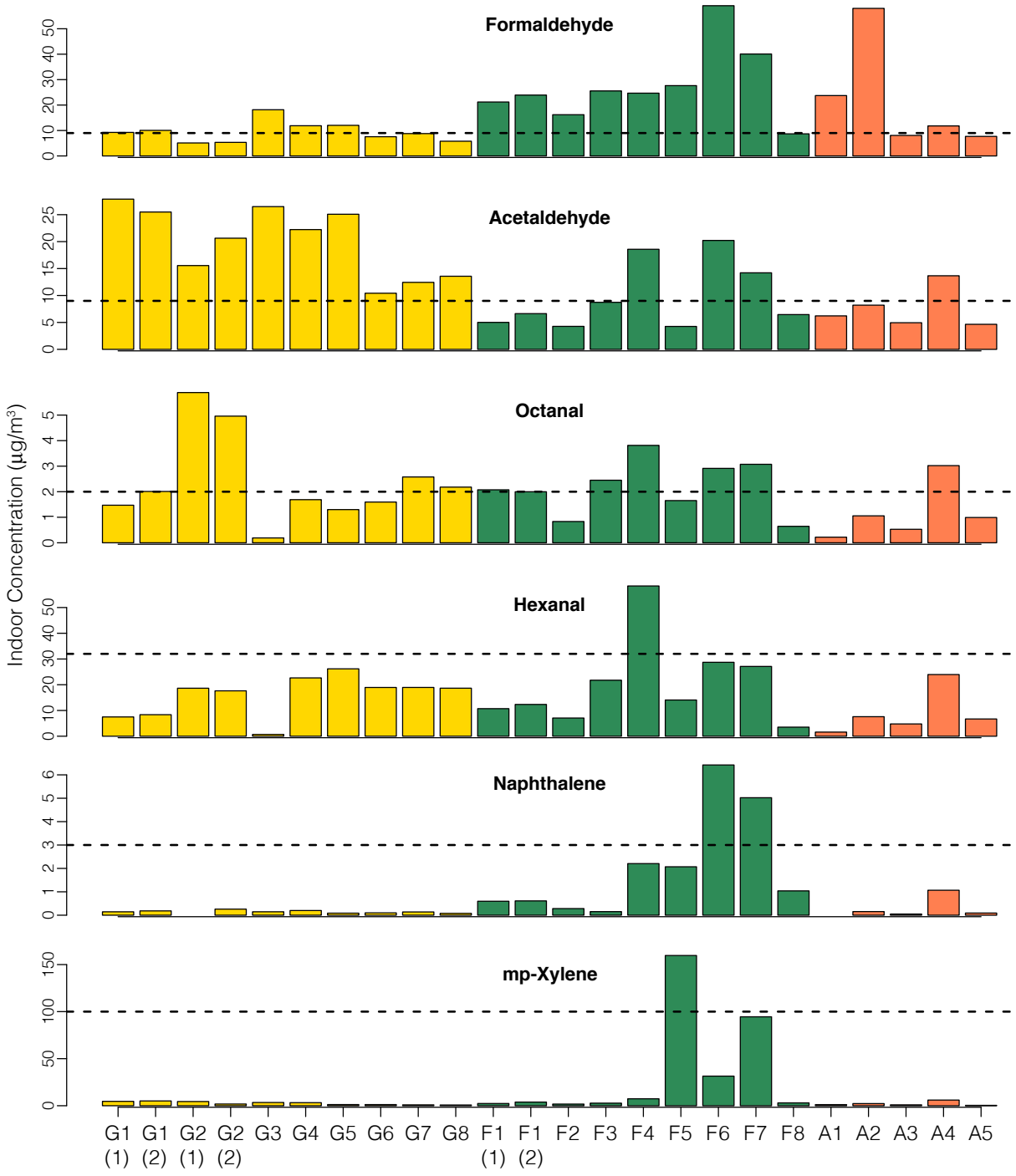

Figure 19 Indoor VOC concentrations measured in three retail store types: grocery (G), furniture/hardware (F), and apparel (A). The dashed line shows the most stringent health guideline or odor threshold (see Table 11). Stores G1, G2, and F1 were sampled on two days: (1) and (2).

Octanal and hexanal are both additives that are used by the food and favoring industry. Octanal has a citrus (orange, grapefruit) flavor. It is found in all three types of retail stores from foods, as well as fragrances used in many consumer products. Grocery stores, such as in store G2, tended to have higher octanal concentrations (Figure 19). Hexanal is described as a grassy scent, such as found in eucalyptus and peppermint oil. It is also commonly found in all three store types, where hardware stores (e.g. stores F4, F6, and F7) with gardening department had the highest concentrations.

Concentrations of naphthalene and m/p-xylene measured indoors were generally low (Figure 19), with higher concentrations in a few of the hardware stores. These two VOCs were found at significant concentrations only in F5, F6, and F7. Off-gassing from deodorizers, repellents, and fumigants has been identified as an important source of naphthalene (Jia, 2010). Xylene is a common component found in solvents being sold in hardware stores.

Other VOCs commonly found in retail stores (see Figure 17) include terpenoids such as d-limonene and a-pinene, texanol used in latex paints, and D5-siloxane used in personal care products, and tetrachloroethylene and 2-butoxyethanol that can be found in solvents used in cleaning products and building materials, and also in merchandise being sold in furniture and hardware stores. The compound 2-ethylhexanol is a fragrance ingredient used in many cosmetics, personal and household products. The highest concentration of 2-ethylhexanol was found in an apparel store A4 (25 $\mu g/m^3$). Store A4 also has the highest concentration of nonanal (18 $\mu g/m^3$), which is also used in fragrances. The other reason why nonanal was commonly found in retail stores is because it is a volatile metabolites emitted from human skin (McGinty, 2010). The typical concentration of nonanal measured in retail stores was about 5 $\mu g/m^3$.

Total VOCs (TVOCs) is a metric used in some green building evaluations, such as the U.S. Green Building Council LEED for New Construction Green Building Rating System (LEED-NC). Given the very wide range in potencies of the VOCs within TVOC to cause adverse health effects, it is unlikely that TVOC is a useful predictor of adverse health effects. LEED-NC gives credit to buildings if indoor air quality testing shows that the TVOC concentration is below 500 $\mu g/m^3$. However, the LEED-NC guidance document (USGBC, 2009) does not specify VOCs that should be included in TVOCs. It also does not specify the sampling procedures and analytical methods that should be used to measure TVOCs. For a rough comparison, in Figure 20, VOCs were grouped into nine classes as they are listed in Table 8, and then they were summed to give the average TVOCs for each type of retail stores.

On average, grocery and apparel stores would easily pass the 500 $\mu g/m^3$ target set by LEED-NC but not the furniture/hardware stores, mostly because of the acetone contained in many home improvement products. Concentrations of aromatics and halogenated compounds were also much more commonly found in furniture/hardware stores than in other two store types. On the other hand, grocery stores tended to have the highest concentrations of terpenoids, such as d-limonene and a-pinene, likely from frequent cleaning that happen in grocery stores. Apparel stores tended to have the highest concentrations of esters because of D5-siloxane, which are commonly found in personal and beauty products.

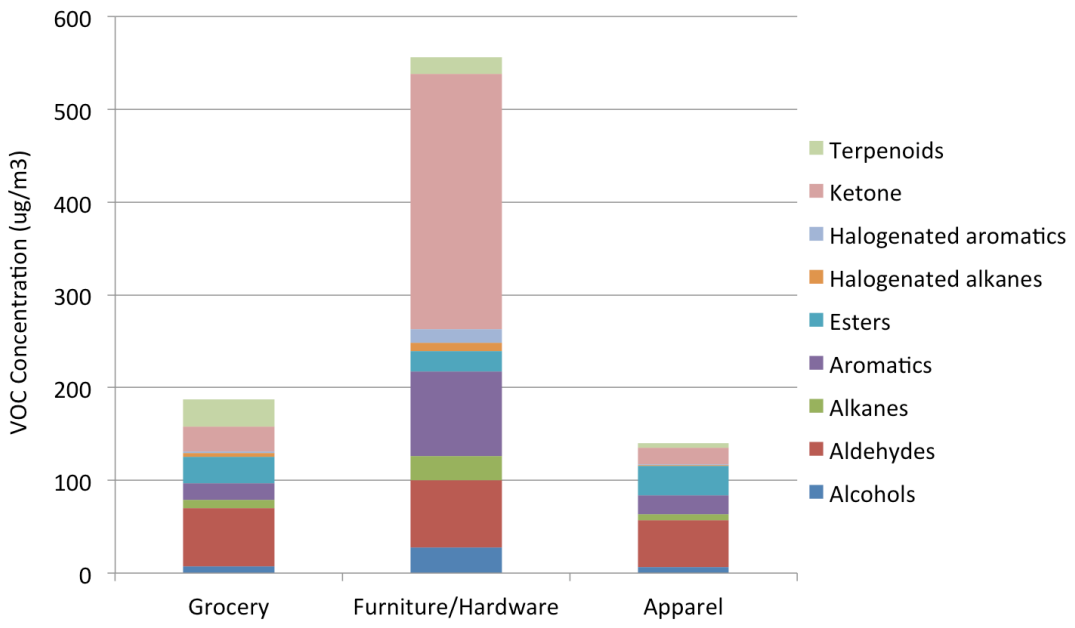

Figure 20 Sum of the average VOC concentrations measured in three types of retail stores. Refer to Table 8 for the list of compounds included in each of the nine classes of chemicals.

Figure 20 shows that acetone is the determinant factor if a retail store would pass the 500 μg/m^3 or not, even though it has relatively low in toxicity compared to other VOCs measured in this study. The Minimal Risk Level set by the Agency for Toxic Substances and Disease Registry is 31 mg/m^3 (ATDSR, 2013) for chronic exposure to acetone, which is three orders of magnitude higher than some of the VOCs that may cause health and odor concerns, such as those listed in Table 8. This analysis shows some of the complexities in evaluating VOC data, which limit the usefulness of TVOCs as a metric for indoor air quality.

5.7 Acrolein

Acrolein may be of concern to human health at very low concentrations. The U.S. EPA (2013) IRIS reference inhalation concentration is 0.02 μg/m^3. The Minimal Risk Levels set by the Agency for Toxic Substances and Disease Registry (ATSDR, 2013) is 0.09 μg/m^3 for intermediate-duration inhalation exposure (15 to 364 days). Environmental tobacco smoke and vehicle exhausts are important sources for exposure to acrolein, as well as the consumption of certain foods (e.g., roasted coffee, fired potatoes). Acrolein is also formed from the heating of fats, which makes it a compound of interests in this study because of cooking in grocery stores. Seaman et al. (2007) found that the increases in acrolein concentrations in residences were correlated with cooking events and air temperature. On average, Seaman et al. (2007) measured 7 μg/m^3 of acrolein in residences using a mist chamber method. The study also found indication of off-gassing and secondary formation of acrolein from sampling conducted in newly built, uninhabited homes. In a related study by Seaman et al. (2009), deep-frying resulted in high concentrations of acrolein in the range of 25–64 μg/m^3 measured a short distance away (6 m) from the source.

Unfortunately, acrolein is difficult to measure because it is highly reactive. Previous field measurements attempted to measure acrolein concentrations in outdoor and indoor air thus far likely underestimated the concentrations because of losses in the sampling method (Ho, 2004). The mist chamber method by Seaman et al. (2007) is not portable. It also takes a long time to set up, making it unsuitable for field measurements. To overcome this problem, a derivatization method to sample acrolein using pentafluorophenyl hydrazine (PFPH)-coated cartridges was adopted based on Ho and Yu (2004), as described below.

5.7.1 PFPH Cartridge Preparation and Sampling

To prepare the PFPH stock solution, 100 mg of PFPH (Sigma Aldrich) was dissolved in 20 mL of acetonitrile. The resulted solution was 5 mg/mL in concentration, and was further diluted by dissolving a 1-mL volume in 39 mL of acetonitrile. The final PFPH stock solution had a concentration of 0.125 mg/mL, and was used in all the analyses described here.

Waters Sep-Pak® silica cartridges were cleaned before use by passing 2 ml of acetonitrile solution through them. They were then dried overnight at 40 ^{o}C, and were kept in the oven until use. One mL of 0.125 mg/mL PFPH solution was passed through the cartridge using a syringe, and the excess was removed from the cartridge using a constant stream of helium for five minutes. The PFPH cartridges were tightly packed in an aluminum foil and stored in a freezer until use.

Acrolein standards were prepared to obtain calibration curves prior to each analysis. 1.8 mL of PFPH was taken in a 2 mL GC vial. To this, 0.2 ml of acrolein standard solution was added, resulting in a total volume of 2 mL. This mixture was allowed to react for a period of 3 hours before it was injected in the GC. A calibration curve was obtained by varying the injection volume. A six-point calibration curve was obtained by injecting 1, 2, 3, 4, 5, and 6 uL to get 66.7, 133.4, 200, 266.8, 333.5, 400 ng of acrolein mass respectively. This calibration curve was used to obtain the mass of acrolein sampled using the cartridges.

Sampling of acrolein was performed initially at a flow rate of 130 mL/min for approximately one hour. The limit of detection was 0.2 ng/m^3. Breakthrough was estimated to occur at ~0.02 mg, which was not an issue for the sampling of acrolein in most indoor environments. Samples were allowed to completely react and form the derivatized compound in room condition for five to seven days before analysis.

Following the sampling in the first two stores (G1 and G2), the sampling flow rate were lowered to 100 mL/min in stores F1–F3 and A1. At the same time, the sampling duration was increased to two hours, so the limit of detection was further lowered to approximately 0.1 ng/m^3. The final set of acroelin measurements made in stores G3–G5 was performed at a lower flow rate of about 80 mL/min. This was done to improve the stability of the sampling flow rates. Sampling duration was lengthened to 2.5 to 3 hours such that the limit of detection was kept at roughly the same level.

Sampling of acrolein concentrations was performed with duplicates using the same equipment as for aldehyde and VOC samples. In grocery stores, acrolein was sampled at four locations. The

sampling locations were the same as where the aldehyde and VOCs samples were collected. In furniture/hardware and apparel stores, acrolein was measured indoors at fewer locations (three in furniture/hardware stores, and one location only in apparel store A1) because concentrations were expected to be low, since there is no clear source of acrolein in those stores.

5.7.2 Results of Acrolein Measurements

Table 13 shows the results of 51 pairs of duplicated samples, including 12 pairs of outdoor samples and 39 pairs of indoor samples (note: two duplicate samples were lost due to sampling error). Duplicate samples tend to agree to 22% (median value) of one another. Overall, the agreement between duplicate samples of acrolein was somewhat lower compared to other VOCs measured using well-established methods of sorbent tubes and DNPH cartridges.

Table 13 Outdoor and indoor acrolein concentrations measured in nine retail stores.

	Outdoor Samples				**Indoor Samples**			
	Start Time	**End Time**	**Flow Rate (mL/min)**	**Acrolein Mean (μg/m^3)**	**Start Time**	**End Time**	**Flow Rate (mL/min)**	**Acrolein (μg/m^3) Mean (Min-Max)**
Grocery Stores								
G1 (1) *	15:28	14:29	145	9.3	15:52	16:55	142	20.5 (7.8–26)
G1 (2) *	14:28	15:29	143	7.3	12:56	13:53	136	9.6 (5.2–12)
G2 (1) *	14:20	15:48	120	4.3	12:27	13:33	116	5.2 (2.8–8.4)
G2 (2) *	13:34	14:33	122	0.4	13:41	14:40	116	4.7 (1.7–6.1)
G3	14:12	17:03	69	1.2	13:52	16:48	88	10.8 (5.5–10)
G4	12:58	15:19	89	0.3	12:39	14:53	76	9.0 (6.8–13.5)
G5	12:52	15:30	66	0.4	12:35	14:50	93	9.0 (4.8–16)
Furniture/ Hardware and Apparel Stores								
F1 (1) *	14:35	16:20	101	0.7	14:10	16:15	108	1.7 (1.6–1.8)
F2 (2) *	13:44	15:44	98	0.8	12:33	14:28	100	1.6 (0.2–3.9)
F2	13:05	15:02	80	0.1	12:59	14:57	115	0.6 (0.05–1.0)
F3	13:40	15:40	76	0.05	12:28	14:26	100	0.3 (0.02–0.6)
A1	13:04	15:03	93	0.1	13:13	15:10	114	3.0 --

* Stores G1, G2, and F1 were measured on two days, as indicated by (1) and (2).

Indoor levels of acrolein in grocery stores were elevated compared to the levels found in other stores, likely because of cooking. On average, approximately 10 μg/m^3 of acrolein was measured in grocery stores (Table 13). Stores that did not have cooking as a source (i.e., stores F1–F3 and A1) had much lower levels of acrolein of about 1 μg/m^3. Still, this level of acrolein found indoors was generally higher than the outdoor level of about 0.5 μg/m^3. This could be an indication of small amount of acrolein being emitted from building materials. High concentrations of acrolein were also measured in some of the outdoor samples. This can be explained by the proximity of the outdoor samplers to the cooking fumes that was being exhausted from the kitchen, and also vehicle emissions that are known sources of acrolein found in ambient air.

This field study was the first that used this particular PFPH method to sample acrolein. Future improvement in the sampling procedure and analytical method can potentially improve the agreement between duplicates. For example, high background level was found in some samples,

possibly because of contamination of the samplers from preparation. In addition, the performance of the method will also need to be more thoroughly evaluated under different environmental conditions.

5.8 Contaminant Source Strengths

5.8.1 Volatile Organic Compounds

Assuming that there is negligible removal of VOCs via mechanisms other than ventilation (i.e., negligible deposition on or reaction with indoor surfaces, no chemical reactions with other indoor air pollutants), the whole-building source strengths of VOCs can be calculated as follows:

$$E\left(\frac{\mu\text{g}}{\text{h}\cdot\text{m}^2}\right)=\frac{\left[C_{in}-C_{out}\right]\left(\frac{\mu\text{g}}{\text{m}^3}\right)\times Q\left(\frac{\text{m}^3}{\text{h}}\right)}{A\left(\text{m}^2\right)}$$

Eq 8

where E is the whole building emission rate per floor area, C_{in} and C_{out} are the indoor and outdoor concentrations, and A is the floor area. The equation also assumes that all parameters are constant, concentrations within the store are well-mixed, and the system has reached steady state.

As shown in Figure 18, there are considerable spatial differences in the VOC concentrations measured inside the stores. For computing E, indoor concentrations were averaged. Indoor concentrations below MDLs were ignored. E was estimated if indoor concentrations were above the MDLs at least at some of the sampling locations, and if the indoor concentrations were above the outdoors. If all the outdoor measurements were below MDLs but an estimate of C_{out} is needed to compute E, it is assumed that the outdoor concentration was negligible (i.e. $C_{out} = 0$).

VOC samples were all collected in the afternoon (see Table 10 for start and end times). Ventilation rates were measured during roughly the same time as the VOC sampling. In mechanically ventilation stores, SF_6 measurements suggest that the ventilation rate stayed more or less constant. Most stores turned off their ventilation system at least partially when they are closed for business. It would take several hours, depending on the air exchange rate, for contaminants to reach a stable level after the store opens. The collection of VOC samples in the afternoon allowed time for indoor concentrations to approach steady-state values.

Figure 21 shows the calculated whole-building VOC emission rates in all retail stores sampled, ordered by the median estimate. Hexanal and acetone are the two VOCs with the highest emission rates found, with the median values approaching 100 μg/h-m^2. They are followed by D5-siloxane, acetaldehyde, d-limonene, and formaldehyde, with emission rates in the range of about 50 μg/h-m^2. There are approximately 15 compounds with an estimated whole building emission rate >10 μg/h-m^2. Below the 10 μg/h-m^2 levels, the emitted VOCs would result in low indoor concentrations that are unlikely to lead to serious health or odor concerns.

For example, assuming an air exchange rate of 0.5 h^{-1}, 4-m store ceiling height, and negligible outdoor concentrations (C_{out} = 0), an emission rate of 10 μg/h-m^2 would give C_{in} = 5 μg/m^3 (Eq 8). Unless the VOCs have health or odor concerns at very low levels (such as acrolein), compounds plotted on the lower half of Figure 21 with E <10 μg/h-m^2 are unlikely to cause health or odor concerns in retail stores.

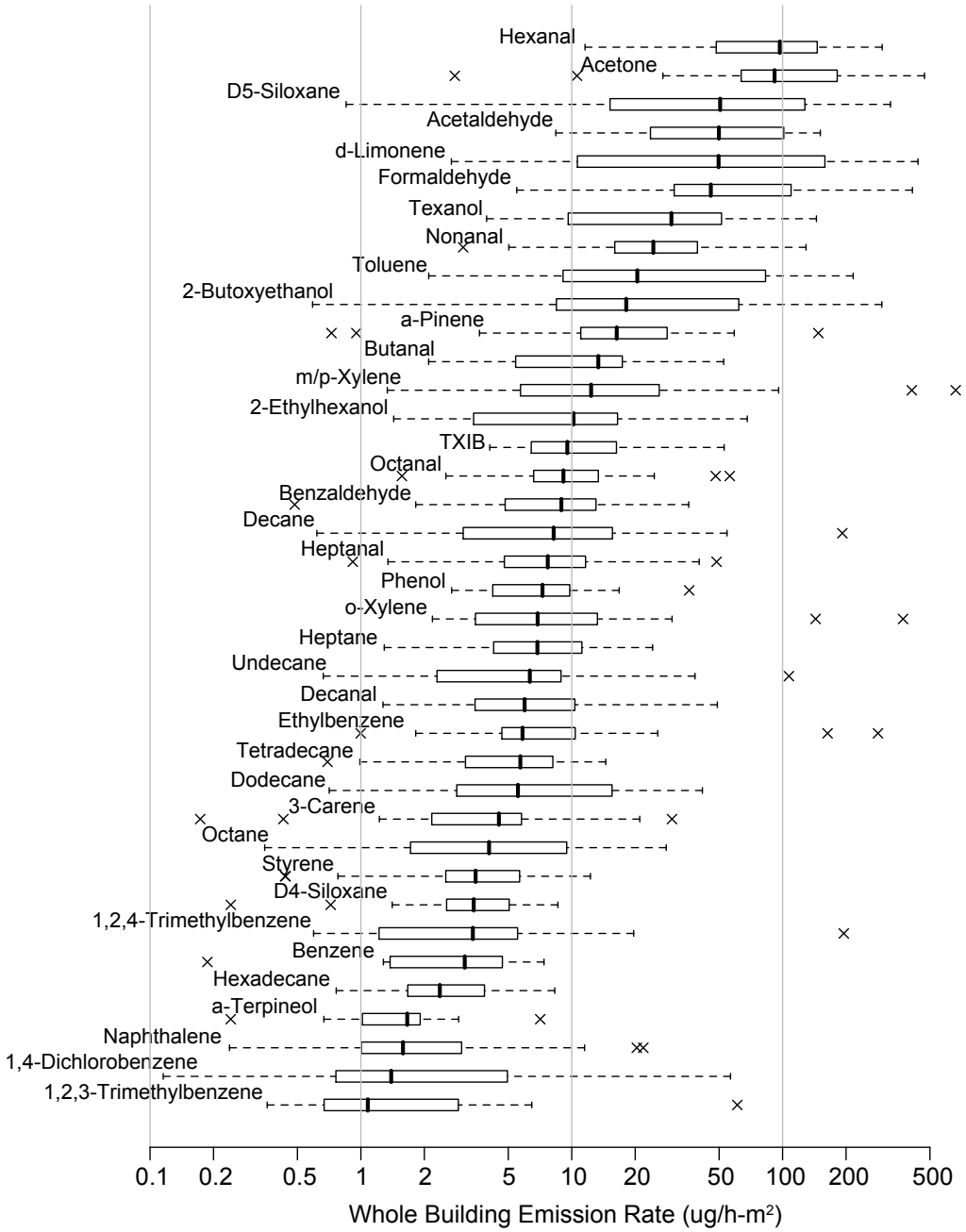

Figure 21 Whole building emission rates of VOCs estimated from concentration measurements collected from 21 retail store visits. Estimates are shown only for VOCs that are frequently measured above MDLs in at least half of the stores. Values outside of the typical range are indicated by the "x" symbols.

The emission rate estimates are usually in rough agreement with the estimates from two other field studies that were recently conducted on retail stores. The first comparison is with a study of small and medium size commercial buildings (SMCB) in California. Wu et al. (2011) estimated the whole-building VOCs emission rates for two grocery stores, five restaurants, and seven retail stores. Table 14 and Table 15 compare the geometric mean (GM) and geometric standard deviation (GSD) of whole-building VOCs emission rates estimates from this study with corresponding data collected by SMCB.

Table 14 Whole building emission rates of high emitting VOCs, in comparison with values reported in SMCB study of grocery stores and restaurants.

	Grocery Stores G1 – G8				**SMCB Grocery/ Restaurants**			
	N	**GM ± GSD**	**Mean ± SD**	**Median**	**N**	**GM ± GSD**	**Mean ± SD**	**Median**
Formaldehyde	9	30 ± 2	35 ± 17	34	8	58 ± 3	90 ± 85	65
Acetaldehyde	9	83 ± 2	102 ± 45	104	8	118 ± 4	216 ± 173	260
Acetone	9	87 ± 2	108 ± 57	94	8	129 ± 9	885 ± 1517	57
d-Limonene	10	176 ± 2	200 ± 109	172	8	56 ± 4	145 ± 213	40
Hexanal	10	121 ± 2	137 ± 59	147	8	25 ± 4	55 ± 91	24
D5-Siloxane	10	96 ± 2	125 ± 101	85	8	124 ± 4	337 ± 563	76
Nonanal	8	37 ± 2	47 ± 37	38	8	42 ± 4	101 ± 186	36
Texanol	8	31 ± 3	47 ± 44	42	8	3 ± 2	3 ± 2	3
a-Pinene	10	26 ± 2	30 ± 18	23	8	10 ± 2	11 ± 6	10
2-Butoxyethanol	8	22 ± 3	38 ± 42	15	8	52 ± 8	294 ± 487	19
Benzaldehyde	10	16 ± 2	19 ± 12	11	4	3 ± 3	4 ± 3	4
Octanal	10	14 ± 2	20 ± 18	12	8	9 ± 3	17 ± 26	8
Toluene	10	13 ± 2	18 ± 16	11	7	7 ± 9	21 ± 28	10
m/p-Xylene	9	11 ± 3	16 ± 11	17	4	3 ± 2	3 ± 2	3
o-Xylene	7	7 ± 2	8 ± 4	7	4	1 ± 2	1 ± .4	1

Table 15 Whole building emission rates of high emitting VOCs, in comparison with values reported in SMCB study of other retail stores.

	Furniture/ Hardware (F1 – F8) and Apparel Stores (A1 – A5)				**SMCB Retail Buildings**			
	N	**GM ± GSD**	**Mean ± SD**	**Median**	**N**	**GM ± GSD**	**Mean ± SD**	**Median**
Formaldehyde	12	85 ± 2	120 ± 13	104	7	88 ± 3	130 ± 129	83
Acetaldehyde	11	28 ± 6	35 ± 105	31	6	22 ± 3	41 ± 63	15
Acetone	11	103 ± 2	337 ± 22	84	6	82 ± 5	229 ± 365	51
d-Limonene	13	14 ± 2	27 ± 107	12	7	34 ± 5	100 ± 170	52
Hexanal	14	57 ± 2	77 ± 71	58	7	12 ± 2	14 ± 8	10
D5-siloxane	14	15 ± 4	58 ± 69	20	7	47 ± 4	78 ± 64	77
Nonanal	12	17 ± 6	22 ± 209	21	6	23 ± 3	41 ± 41	24
Texanol	14	19 ± 3	35 ± 37	20	7	7 ± 2	9 ± 5	7
a-Pinene	14	10 ± 8	22 ± 81	13	7	11 ± 6	34 ± 47	13
2-Butoxyethanol	13	22 ± 3	73 ± 35	21	7	9 ± 6	27 ± 43	12
Benzaldehyde	14	5 ± 5	8 ± 113	6	4	2 ± 3	3 ± 2	3
Octanal	14	7 ± 2	9 ± 13	9	7	9 ± 5	38 ± 80	6
Toluene	12	40 ± 4	77 ± 37	66	6	34 ± 5	81 ± 112	33
m/p-Xylene	12	21 ± 5	106 ± 53	12	6	5 ± 3	7 ± 4	7
o-Xylene	11	12 ± 3	54 ± 6	6	6	3 ± 4	5 ± 3	5

The agreement with the SMCB study is particularly good among the retail stores summarized in Table 15. For example, formaldehyde whole building emission rate estimated from F1–F8 and A1–A5 is 85±2 μg/h-m^2 (GM ± GSD). SMCB found the formaldehyde emission rate to be 88±3 μg/h-m^2. For acetaldehyde, the estimated emission rate from this study is 28±6 μg/h-m^2, and 22±3 μg/h-m^2 from SMCB. All other VOCs emission rates tabulated in Table 15 have roughly the same order of magnitude.

There are more differences between the grocery stores sampled in this study and the grocery/restaurants sampled by SMCB (Table 14). This is because the SMCB data shown was mostly from restaurants, and not grocery stores. Emission rates of VOCs associated with cooking and baking, such as acetaldehyde, acetone, and 2-butoxylethanol, were substantially higher in restaurants sampled by SMCB. The grocery stores measured in this current study also tend to be larger in floor area. Less surface area-to-volume ratio would explain the lower formaldehyde emission rates 30±2 μg/h-m^2 estimated in this study, relative to 58±3 μg/h-m^2 estimated by SMCB, for example.

The ASHRAE Project 15960-RP (Siegel, 2012) measured formaldehyde, acetaldehyde, and other VOCs in fourteen big-box stores. They include two home improvement stores, two furniture stores, four grocery stores, four general merchandise stores, one electronic store, and one office supply store. Figure 22 compares the emission rates of formaldehyde and acetaldehyde measured in this study with the estimates measured by the ASHRAE project and by SMCB.

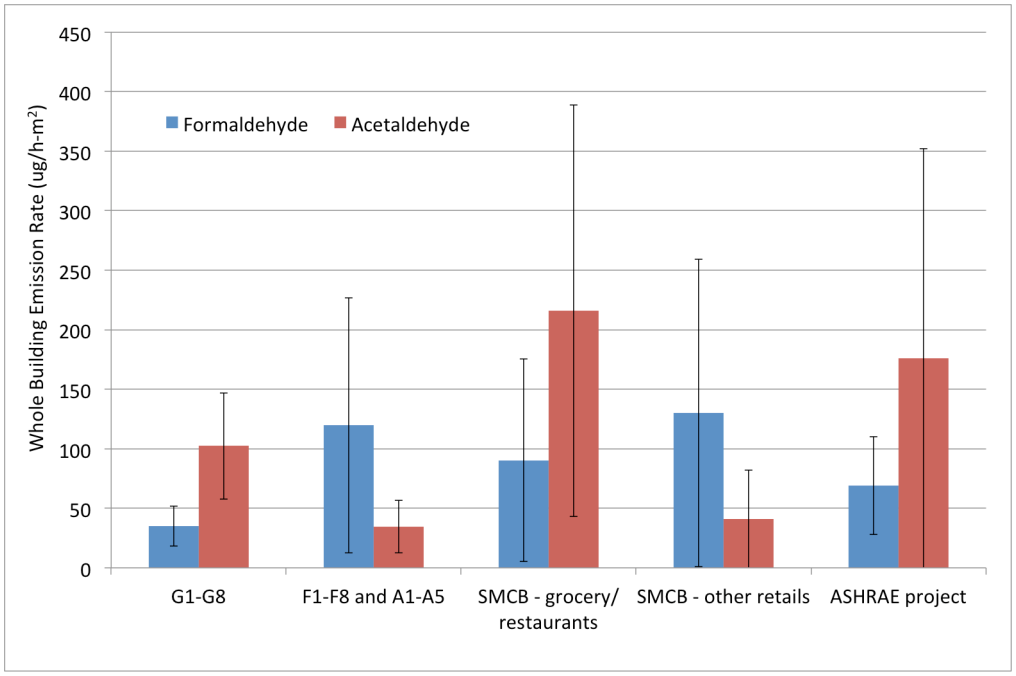

Figure 22 Comparison of formaldehyde and acetaldehyde whole building emission rates estimated from this study, with estimates from SMCB (Wu et al. 2011) and ASHRAE project (Siegel et al. 2012). The average emission rate estimated from the sampled stores is plotted with the standard deviation indicated by the error bar.

The formaldehyde emission rates measured by the ASHRAE project fall between the lower values estimated from grocery stores (G1–G8), and the higher values estimated from furniture/hardware (F1–F8) and apparel (A1–A5) stores. The emission rates of acetaldehyde estimated from this study are lower than the ASHRAE project. The ASHRAE project found elevated concentrations of acetaldehyde not only from grocery stores, but also from one home improvement and one general merchandise store. Siegel et al. (2012) explained that acetaldehyde measured may be produced by combustion, and from building materials and consumer products such as adhesives, coatings, lubricants, and inks. However, this study only found significant sources of acetaldehyde from grocery stores, and not in other store types. Both this study and SMCB found a similar pattern of high acetaldehyde and relatively lower formaldehyde in grocery stores, and the reverse to be the case in other store types (Figure 22).

Whole building emission rates can vary quite substantially from day to day. Table 16 shows that in the three stores (G1, G2, and F2) that were measured on two consecutive days, emission rates of VOCs tend to vary by about 35%. In two stores that were measured in two different seasons (G3 in May 2012 and February 2013, and F6 in September 2012 and February 2013), the difference in VOC emission rates was about 55%. In comparison to these emission rate estimates, ventilation rates did not change very much in stores, especially if they were ventilated mechanically.

Table 16 Comparison of VOC emission rates and ventilation rates measured in five stores where samples were collected on two days.

	Stores sampled on two consecutive days			**Stores measured on two days that were a few months apart**	
VOC [a]	**G1(1) and (2)**	**G2(1) and (2)**	**F1(1) and (2)**	**G3 and G7**	**F6 and F7**
Formaldehyde	59%			13%	15%
Acetaldehyde	22%			65%	8%
Acetone	11%			15%	135%
d-Limonene	2%	39%	40%	37%	90%
Hexanal	9%	9%	2%	28%	22%
D5-Siloxane	0.3%	15%	45%	44%	54%
Nonanal	26%			98%	11%
Texanol	54%	15%	47%	81%	63%
a-Pinene	45%	50%	20%	2%	73%
2-Butoxyethanol	9%	21%	14%	70%	23%
Benzaldehyde	5%	5%	8%	36%	37%
Octanal	35%	15%	19%	78%	33%
Toluene	175%	11%		29%	18%
m/p-Xylene		66%	102%	158%	124%
o-Xylene		110%	67%		131%
Average	35%	32%	36%	54%	56%
Range (Min-Max)	0.3%–175%	5%–110%	2%–102%	2%–158%	8%–135%
ACH (h^{-1})	5% (Mech. Vent.)	3% (Mech. Vent.)	20% (Nat. Vent.)	16% (Mech. Vent.)	32% (Mech. Vent.)

[a] Only emission rates with store averaged value >1 $\mu g/h\text{-}m^2$ are shown in this table.

The ASHRAE project reported a relative percent variance of 44±39% for formaldehyde emission rates, and 76±58% for acetaldehyde, from stores that were sampled on multiple visits. These temporal differences are similar in magnitude (about 55% on average) to the estimates shown in

Table 16. From this, Siegel et al. (2012) concluded that indoor activities and sources differed between visits. Also similar to findings from this study is that air exchange rate varied less between visits. ASHRAE project reported that air exchange rate varied by 22±22%.

This current study further observed that compounds such as toluene and xylenes had estimated emission rates that varied the most temporally (see Table 16). Therefore, the characterization of indoor concentrations, and subsequent estimates of the emission rates of these compounds, will require more extensive sampling to quantify. On the other hand, compounds such as hexanal and benzaldehyde, both are compounds used as flavoring agents and fragrances, stayed relatively constant likely because they are ubiquitous in products being sold in stores.

5.8.2 Acrolein

Grocery stores that were sampled in this study clearly had sources of acrolein. Using Eq 8, sampling from five grocery stores suggests an emission rate of about 45 μg/h-m^2 (Table 17).

Table 17 Estimates of acrolein emission rates.

Grocery Stores	**E (μg/h-m^2)**	**Furniture/ Hardware Stores**	**E (μg/h-m^2)**
G1	62 (day 1) 14 (day 2)	F1	5 (day 1) 3 (day 2)
G2	9 (day 1) 41 (day 2)	F2	7
G3	76	F3	0.4
G4	45	**Apparel Store**	
G5	38	A1	21
Average =	45	Average =	8

The estimated acrolein source strength of 45 μg/h-m^2 in grocery stores is higher than that of formaldehyde, where the mean source strength estimated for grocery stores is 35 μg/h-m^2 (Table 14). Because of the very low chronic health guideline of acrolein (0.02 μg/m^3 (U.S. EPA) or 0.09 μg/m^3 (ATSDR), i.e. approximately an order of magnitude less than that of formaldehyde), there may be substantial health risks associated with this exposure. In other stores without cooking, the average emission rate was about 8 μg/h-m^2 (Table 17). As a comparison, Seaman et al. (2007) estimated an average emission rate of 5 μg/h-m^2 based on measurements of acrolein concentrations collected once in the morning and once in the evening in nine homes. Higher acrolein concentrations were measured in homes with frequent cooking, and also in the evenings when cooking tends to occur.

In stores G1 and G2, substantial differences were found between the acrolein emission rate estimated from measurements from two consecutive sampling days. This may be an indication that concentrations in the stores were changing in response to the level of cooking activities in the stores. Anecdotally, cooking occurred throughout the day in grocery stores. In both G1 and G2, estimates of acrolein emission rates were lower on the day when sampling occurred earlier in the afternoon (refer to Table 13 for sampling times). This may be an indication that concentrations of acrolein were building up inside the grocery stores, as cooking fumes accumulated.

In comparison, concentrations measured in store F1 were very similar on both days, even though sampling occurred much earlier in the afternoon on day 2 than day 1. In stores G3–G5, sampling durations were lengthened from about one hour to two or three hours. The longer sampling time likely improved the representativeness of the measured acrolein concentrations and also the subsequent estimates of the emission rates.

5.8.3 Particulate Matter

Whole-building emission rates of PM2.5 and PM10 were estimated. However, many assumptions were required and the uncertainty in the resulting estimates is high. Emission rates were estimated as follows:

$$E\left(\frac{\mu g}{h \cdot m^2}\right) = \frac{\left\{C_{in}\left[\eta \cdot k_{rec} + k_{inf} + k_{dep} + k_{oa}\right] - C_{out}\left[P \cdot k_{inf} + k_{oa}\left(1-\eta\right)\right]\right\}\left(\frac{\mu g}{h \cdot m^3}\right) \times V\left(m^3\right)}{A\left(m^2\right)}$$

Eq 9

where C_{in} and C_{out} are the indoor and outdoor PM2.5 and PM10 mass concentrations, as shown in Table 6. Other variables that were used to estimate the particle emission rates are defined subsequently in Table 18.

In Eq 9, the measured air exchange rate shown in Table 2 was separated into two parts for mechanically ventilated stores: infiltration k_{inf} (h^{-1}) that occurred mostly through large openings such as the store entrance, and outside air intake k_{oa} (h^{-1}) through the mechanical rooftop unit. Siegel et al. (2012) found that infiltration accounts for 50% of the air exchange in two grocery stores (a smaller grocery store had infiltration accounting for >100% of the air exchange, likely due to measurement error). Most of the air infiltrated into the building through the store entrance doors (Siegel, 2012). As a result, a penetration factor $P = 1$ is assumed (i.e. minimal loss in particles as they enter through the store entrance doors) for all retail stores.

Siegel et al. (2012) also found that infiltration was often negligible at the other retail stores measured, including a hardware store, an electronic store, an office supply store, and two furniture stores. But on one visit at two of the five stores listed above, infiltration was estimated to account for 40% and 60% of the air exchange. Overall, the data suggests that air infiltration rates in many retail stores were likely to be low. For this study, it is assumed that furniture/hardware and apparel stores had lower infiltration rates than grocery stores because fewer shoppers visited the stores during sampling, meaning there was likely less air infiltration from opening and closing of the store doors. For the furniture/hardware and apparel stores that were mechanically ventilated, air infiltration was assumed to equal 10% of the measured air exchange rate.

Siegel et al. (2012) measured the outdoor air percentage in retail stores by measuring the CO_2 concentrations in the outside air, supply air, and recirculated air. The measurements from seven retail stores showed that on average, about 25% of the total supply air flow was from outside air, and the remaining is recirculation. Thus in Table 18, k_{rec} is assumed to equal ×3 the measured air

exchange rate, based on the ratio of recirculated air to outdoor air measured by Siegel et al. (2012).

The first-order particle deposition loss rates, k_{dep} (h^{-1}), and the particle removal efficiencies of filters, η, are based on modeling results by Riley et al. (2002). That study used models with experimentally validated data to predict particle deposition loss coefficients and particle removal efficiencies of air filters. The estimates took into account particle size distributions for PM2.5 and PM10, and also the tendency of filters to preferentially remove particles that deposit on surfaces the most readily. From Riley et al. (2002), deposition coefficient k_{dep} of 0.13 h^{-1} for PM2.5 and 0.54 h^{-1} for PM10 were used in Table 18.

The removal efficiency of various air filters in reducing PM2.5 and PM10 were determined based on the analysis by Riley et al. (2002). Riley et al. (2002) estimated the particle removal efficiency of two air filters that had an efficiency rating approximately equivalent to MERV 8 and MERV 13. To account for improved efficiency with increasing filter loading (Hanley, 1994), filter efficiencies estimated per Riley et al. (2002) were adjusted to η = 18% (PM2.5) and 24% (PM10) for MERV 8, and η = 70% (PM2.5) and 74% (PM10) for MERV 13 filters. Store G1 used MERV 15 bag filters. Fisk et al. (2002) estimated the particle removal efficiency curve for such bag filters to be slightly better than ASHRAE 85% air filters. But the overall difference in efficiency is small between the MERV 15 bag filters and MERV 13 pleated air filters, so it is assumed that the same values of η used to describe MERV 13 air filters also applied for store G1.

Table 18 shows the values of the input parameters to Eq 9, and the resulted PM2.5 and PM10 emission rates. Emission rates were substantially higher in grocery stores than in furniture/hardware and apparel stores for both PM2.5 and PM10.

In naturally ventilated stores, there is no mechanically supplied outside air, so k_{inf} simply equals the measured air exchange rate. There is also no air filter to remove particles. In such stores, the whole-building PM2.5 and PM10 emission rates were largely determined by k_{dep}. In some cases such as in stores F1(1), F2, F5, and A2, it is necessary to adjust k_{dep} to higher values in naturally ventilated stores such that the estimated PM2.5 emission rate is not a negative number. The minimum values of k_{dep} that give non-negative estimates of E are shown in Table 18.

In addition to the adjustments made to k_{dep}, it was also necessary to adjust the filter efficiency η in stores F4, F6, F8, and A4, such that the calculations did not give negative estimates of emission rates. Store F6 used MERV 2 filters that were assumed to have negligible efficiency. Table 18 suggests that the filter efficiency may have been somewhat higher at 5% to 10% range, perhaps due to heavy particle loading leading to an increase in removal efficiency. Stores F4, F8 and A4 used MERV 8 filters. The estimated efficiencies from this analysis were also higher than initially assumed.

Table 18 Estimates of PM2.5 and PM10 emission rates.

	k_{rec}	k_{inf}	k_{oa}	k_{dep}		η		E (μg/h-m^2)	
				PM2.5	**PM10**	**PM2.5**	**PM10**	**PM2.5**	**PM10**
Grocery Stores									
G1 (1)	2.2	0.4	0.4	0.13	0.54	0.70	0.74	105	156
G1 (2)	2.3	0.4	0.4	0.13	0.54	0.70	0.74	47	129
G2 (1)	4.4	0.7	0.7	0.13	0.54	0.18	0.24	135	166
G2 (2)	4.3	0.7	0.7	0.13	0.54	0.18	0.24	157	313
G3	2.3	0.4	0.4	0.13	0.54	0.18	0.24	318	696
G4	2.2	0.4	0.4	0.13	0.54	0.18	0.24	112	143
G5	2.0	0.3	0.3	0.13	0.54	0.18	0.24	33	80
G6	3.4	0.6	0.6	0.13	0.54	0	0	126	188
G7	2.0	0.3	0.3	0.13	0.54	0.18	0.24	211	346
G8	6.0	1.0	1.0	0.13	0.54	0.18	0.24	254	571
Furniture/ Hardware Stores									
F1 (1)	--	1.2	--	**0.17** [a]	0.54	--	--	**NA** [a]	3
F2 (2)	--	1.0	--	0.13	0.54	--	--	6	2
F2	--	2.4	--	**0.27** [a]	0.54	--	--	**NA** [a]	76
F3	--	0.4	--	0.13	0.54	--	--	3	27
F4	2.0	0.1	0.6	0.13	0.54	**0.20** [b]	0.24	**NA** [b]	80
F5	--	1.2	--	**0.64** [a]	0.54	--	--	**NA** [a]	26
F6	1.6	0.1	0.5	0.13	0.54	**0.09** [b]	**0.06** [b]	**NA** [b]	**NA** [b]
F7	2.2	0.1	0.7	0.13	0.54	0	0	31	130
F8	3.5	0.1	1.0	0.13	0.54	**0.33** [b]	0.24	**NA** [b]	30
Apparel Stores									
A1	--	2.3	--	0.13	0.54	--	--	9	87
A2	--	2.2	--	**0.20** [a]	0.54	--	--	**NA** [a]	23
A3	1.5	0.1	0.5	0.13	0.54	0.18	0.24	18	110
A4	1.6	0.1	0.5	0.13	0.54	**0.40** [b]	**0.40** [b]	**NA** [b]	**NA** [b]
A5	6.6	0.2	2.0	0.13	0.54	0	0	6	63

[a] Minimum deposition coefficient to give a non-negative estimate of emission rate, *E*.
[b] Minimum filter efficiency to give a non-negative estimate of *E*.

Excluding cases where either the deposition coefficient or the filter efficiency had to be adjusted in order to give a sensible estimate of the whole-building PM2.5 and PM10 emission rate, Figure 23 shows the average emission rates (minimum and maximum showed as error bars) by store types.

Grocery stores had indoor emissions of PM2.5 and PM10 that averaged about 150 and 280 μg/h-m^2, respectively. Furniture/hardware and apparel stores had negligible sources of PM2.5 that only averaged to 12 μg/h-m^2. Indoor sources of PM10 were also small in these two store types, where the average emission rate of PM10 was 55 μg/h-m^2.

In comparison to findings from the ASHRAE project (Siegel, 2012), this study found higher emissions of PM2.5 in grocery stores. Emission estimates of PM10 from this study and by the ASHRAE project are within the same range in grocery stores. Both the ASHRAE project and this study estimated lower emission rates of PM2.5 and PM10 for the non-grocery stores.

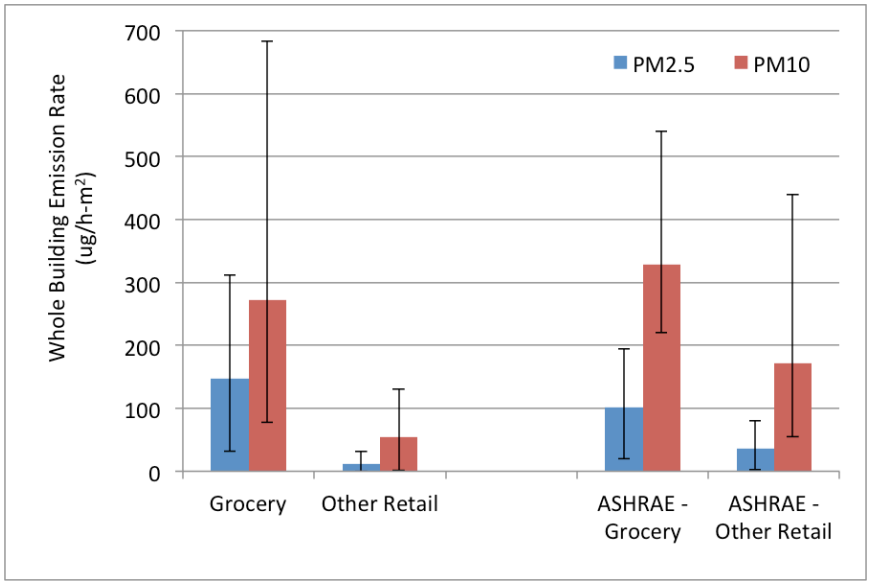

Figure 23 Estimated whole building emission rates of PM2.5 and PM10 in grocery stores, and in the other retail types. Emission rates estimates are compared with values reported by the ASHRAE project. Vertical bars plotted show the average emission rates. Error bars show the range of estimates (minimum and maximum).

From the same equation used to estimate whole building emission rates (Eq 9), the percentage contribution of indoor sources to the indoor PM concentrations can be computed as follows:

$$\text{Indoor Contribution } (\%) = \frac{1}{1 + \dfrac{C_{out}\left[P \cdot k_{inf} + k_{oa}\left(1-\eta\right)\right]\left(\dfrac{\mu g}{h \cdot m^3}\right)}{E\left(\dfrac{\mu g}{h \cdot m^2}\right)\dfrac{A\left(m^2\right)}{V\left(m^3\right)}}}$$

Eq 10

Figure 24 shows that indoor sources contributed about 70% of the indoor PM2.5 measured in grocery stores. This is similar to estimates by the ASHRAE project from measurements of two grocery stores.

In this study, furniture/hardware and apparel stores did not have significant indoor sources of PM2.5. Estimates of indoor contributions were about 30% in stores that kept their entrance door closed (i.e. all mechanically ventilated stores, plus F3 which was naturally ventilated but chose to keep entrance door closed).

Stores that rely on natural ventilation tend to leave their entrance door open. Consequently, naturally ventilated stores are highly impacted by outdoor PM levels. The parameters used in this analysis resulted in about 90% of the indoor PM2.5, and 85% of indoor PM10, being the direct consequences of particles that originated from outdoor air.

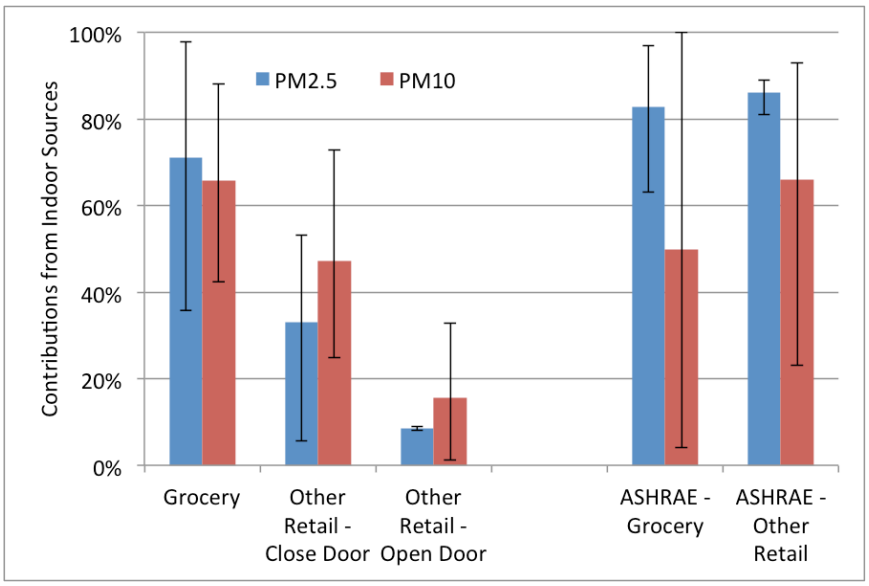

Figure 24 Contribution estimates of indoor sources to PM2.5 and PM10 in grocery stores, and in other retail stores that kept the entrance door opened (F1, F2, F5, A1, and A2), and in other stores that kept it closed (F3, F4, F6–F8, A3–A5). Predictions are compared with values estimated by the ASHRAE project. Vertical bars plotted show the average emission rates. Error bars show the range of estimates (minimum and maximum).

There are two key differences between the estimates from the ASHRAE project and this study. The first is that the ASHRAE project sampled stores that are larger in size. Many of them also sell a wider range of merchandises. Assuming that these store characteristics implied more indoor activities that may generate or resuspend particles, this would explain why higher contributions from indoor sources were estimated by the ASHRAE project, compared to this study. Relative to this study, the ASHRAE project also found more variability in the source estimates, particular in the case of indoor contributions of PM10.

The second difference is that this study found relatively higher indoor source contributions to PM10 than to PM2.5 in furniture/hardware and apparel stores. On the other hand, the ASHRAE project found the opposite to be true. The "other retail" sampled in this study represent stores that had very low indoor PM sources, while the ASHRAE project included some stores in the "other retail" category that had significant indoor sources. For example, an electronics store sampled by the ASHRAE project had the second highest PM2.5 emission rates after grocery stores. Consequently, the ASHRAE project found indoor sources contributing to about 85% of PM2.5 measured indoor in both grocery and other retail stores, as shown in Figure 24. The ASHRAE project reported highly variable proportions of PM10 emitted from indoor sources, but on average, contributions from indoor sources estimated for PM10 are less than that for PM2.5.

6 Implications to Title 24 Ventilation Standards

The Title 24 ventilation requirement was met in all thirteen stores with mechanical ventilation. Five of the six stores that relied on natural ventilation at least for most of the times also had sufficient ventilation per Title 24. Overall, retail stores sampled had an average air change rate of 1 h^{-1}. The ventilation requirement of Title 24 is roughly equivalent to 0.5 h^{-1} among grocery stores, and 0.75 h^{-1} among the other two store types (furniture/hardware and apparel) that were sampled. The difference between these two Title 24-equivalent air change rates is due to taller ceiling heights in grocery stores (about 7.5 m) than in the other two store types (about 5.5 m).

Even though retail stores tend to operate at higher ventilation rates than required by Title 24, formaldehyde remains a concern when compared with the most stringent chronic health guidelines. In grocery stores where cooking occurred, acetaldehyde and acrolein were the contaminants that also demand better controls. Cooking likely led to higher particle levels in grocery stores. Finally, this study included several stores that relied on natural ventilation for the majority of the time. For them, the main concern is outdoor particles entering into the stores. The implications of each of these three issues with regard to Title 24 are discussed below.

6.1 Stores with Formaldehyde being the Key Contaminant of Concern

In order to maintain the formaldehyde levels in retail stores to below the OEHHA guideline of 9 $\mu g/m^3$, many of the stores sampled would need to do one or more of the following: (i) reduce their formaldehyde emissions by a substantial amount, (ii) dramatically increase ventilation rates from their current levels that already exceeded Title 24 requirements, or (iii) implement effective air cleaning to remove formaldehyde from the indoor air. In particular, furniture/hardware stores would need to cut emissions by an average of 70% to meet this most stringent health guideline.

If ventilation rates could be increased to lower formaldehyde indoors, there would be a modest reduction in cancer risks from this exposure. OEHHA (2011) published cancer potency factor for formaldehyde and other air toxics. The cancer risk is 6 potential excess cancers among a million people continuously exposed to 1 $\mu g/m^3$ of formaldehyde over 70-year lifetime. There are nearly 200,000 employees working in furniture and hardware stores in California from the 2007 Economic Census data (Census, 2009). In furniture and hardware stores, workers are typically exposed to about 30 $\mu g/m^3$ of formaldehyde. Assuming a 40-hour workweek and exposure duration of 40 years, the potential excess cancer cases is about 5. If furniture/hardware stores were to increase the ventilation rate from their current 1 h^{-1} to 1.5 h^{-1} (which this is about twice the minimum requirement of Title 24), indoor formaldehyde concentrations would be expected to decrease from 30 $\mu g/m^3$ to about 20 $\mu g/m^3$. This calculation assumes an average outdoor formaldehyde concentration of 4 $\mu g/m^3$ as observed in this study. Consequently, the excess cancer cases are expected to lower from 5 to 3.5 among workers.

For shoppers who spend on average 30 minutes per week shopping in malls and stores (NHAPS data (Klepeis, 2001)), assuming that one out of five trips is to either a furniture or hardware store, then the excess cancer risk over a 70-year lifetime for the population of 38 million Californians is 4. If furniture/hardware stores were to increase ventilation rate from their current

1 h^{-1} to 1.5 h^{-1} such that the indoor formaldehyde concentrations would decrease from 30 $\mu g/m^3$ to about 20 $\mu g/m^3$, the excess cancer cases are expected to lower from 4 to 3 among shoppers.

Given that the effects of ventilation rate on cancer risk and other chronic health effects are relatively small, Mendell and Fisk (2013) argue that minimum ventilation requirements, at least in office buildings, should be based on the need to prevent acute outcomes. These may include building related sick building syndrome symptoms, dissatisfaction with perceived air quality, and decrements in work performance. Mendell and Fisk (2013) further identified areas that demand more data: notable are the relationship of ventilation rates to illness absence rates, and also to the rates of communicable respiratory infections. Some of these concerns are also relevant in retail stores.

A detail energy cost analysis is outside the scope of this report, but one can expect that the cost of raising Title 24 minimum ventilation requirements such that the 15,000 furniture and hardware stores would ventilate at 50% above the current requirement is likely to be significant. Consequently, it is doubtful if more ventilation is the most suitable option to lower formaldehyde levels in all retail stores. In addition, this study points out the large variability in source strengths. Even among stores of the same type, this variability between stores indicates that raising the minimum ventilation rate requirement might not be the best solution to reduce formaldehyde exposures.

Overall, formaldehyde levels measured in this study were lower than the study by Wu et al. (2011) where sampling took place between 2009 and 2010 in California small and medium commercial buildings. Formaldehyde levels were also lower than the ASHRAE project conducted between 2011 and 2012 in Texas and Pennsylvania (Siegel, 2012). These comparisons suggest that adoption of the California ARB's Composite Wood Products Airborne Toxic Control Measure on formaldehyde emissions may have been helpful in mitigating this potential concern in retail stores. Some examples of success in this study include furniture store F3 and hardware store F8, where the estimated emission rates were about 30 $\mu g/h\text{-}m^2$. At this low level, the resulted indoor concentration would meet the most stringent chronic health guideline of 9 $\mu g/m^3$ at the current Title 24 ventilation rate. Very low levels of formaldehyde emissions were also measured in two recently renovated apparel stores A3 and A4. These two stores belong to the same retail chain that used low emitting materials in their recent renovations, thus, the data suggest that source control can be an effective strategy.

6.2 Grocery Stores with Cooking Emissions

This study shows that grocery stores have significant emissions of acetaldehyde, acrolein, and particles, all of which are likely emitted from cooking, baking, and other food preparation activities. The following analyses use typical emission rates of these contaminants estimated from this study to approximate indoor concentrations that may result. While the data collected from this study may not reflect the full range of store conditions because of small sample size, it is expected the data describe the usual levels of contaminants that are commonly present. The typical whole building emission rate of acetaldehyde is about 100 $\mu g/h\text{-}m^2$ from the seven grocery stores measured. At an air exchange rate of 1 h^{-1} and ceiling height of 7.5 m, the

resulting indoor acetaldehyde concentration is 13 μg/m^3. This exceeds the most stringent acetaldehyde U.S. EPA health guideline of 9 μg/m^3, but it is far lower than California OEHHA level of 130 μg/m^3.

In comparison, acrolein is the contaminant with indoor concentrations that far exceeded all health guidelines. In grocery stores, typical emission rate is 45 μg/h-m^2. This means an hourly emission rate of 135 mg/h in a 3,000 m^2 store, which is very significant. To put this into perspective, acrolein emissions from a cigarette is 0.1 to 2 mg (U.S.EPA, 2003), meaning that the acrolein emission rates estimated in grocery stores is equivalent to the acrolein emissions from about 100 cigarettes smoked indoors every hour. Even in stores that do not have cooking as an indoor source, the whole building emission rate is not negligible, but it does tend to be lower at about 8 μg/h-m^2. If better capture efficiency by kitchen exhaust is a viable way to lower the acrolein concentrations in grocery stores, then a reasonable target might be to aim at reducing the whole building emission rates to a level that is more commonly found in stores without cooking. For example, if emissions could be reduced to 10 μg/h-m^2 by improving the effectiveness of kitchen exhaust, acrolein concentrations in grocery stores could be lowered to 1 μg/m^3, which is the level that are more commonly observed in other retail stores without cooking.

Cooking is also a source of particle emissions. In grocery stores, emission rates are estimated to be about 150 μg/h-m^2 for PM2.5, and 280 μg/h-m^2 for PM10. Assuming a typical air exchange rate of 1 h^{-1} found in grocery stores sampled, of which half entered the store via the mechanical system, and the other half of outside air entered into the store via entrance door, and using the same recirculated air flow rate and the particle deposition rate as the previous analyses, then using MERV 13 air filters instead of MERV 8 can reduce the indoor PM levels by approximately 50%. Table 19 shows the estimated indoor PM2.5 and PM10 concentrations for stores with typical and high indoor emissions, and when the outdoor PM levels are typical or higher than usual. The fractional improvement going from MERV 8 to MERV 13 depends slightly on the outdoor concentrations and the estimated indoor emission rates.

Table 19 Predicted indoor PM2.5 and PM10 in grocery stores with different efficiency air filters.

	Outdoor		**Indoor w/MERV 8 Filters**		**Indoor w/MERV 13 Filters**	
Typical indoor emissions: 150 μg/h-m^2 (PM2.5) and 280 μg/h-m^2 (PM10)						
	PM2.5 (μg/m^3)	**PM10 (μg/m^3)**	**PM2.5 (μg/m^3)**	**PM10 (μg/m^3)**	**PM2.5 (μg/m^3)**	**PM10 (μg/m^3)**
Typical Outdoor PM	10	20	18	25	8	14
High Outdoor PM	35	50	32	37	14	19
High indoor emissions: 300 μg/h-m^2 (PM2.5) and 540 μg/h-m^2 (PM10)						
	PM2.5 (μg/m^3)	**PM10 (μg/m^3)**	**PM2.5 (μg/m^3)**	**PM10 (μg/m^3)**	**PM2.5 (μg/m^3)**	**PM10 (μg/m^3)**
Typical Outdoor PM	10	20	30	40	15	23
High Outdoor PM	35	50	44	52	20	28

Table 19 shows the benefit of using higher efficiency air filters. Using the California annual PM2.5 standard of 12 $\mu g/m^3$ and annual PM10 standard of 20 $\mu g/m^3$ as the health guidelines, MERV 13 air filters are needed in all grocery stores. MERV 8 air filters may be insufficient to maintain indoor PM to levels that are considered as protective for public health. In grocery stores with very high emission rates (modeled as twice the "typical" values in Table 19), even MERV 13 air filters would not be sufficient to maintain indoor PM to levels that meet the annual ambient air quality standards. Grocery stores with very high whole-building emission rates of PM2.5 and PM10 would likely benefit from using filters with higher MERV ratings than the commonly used MERV 8 pleated filters. The indoor air quality in such stores would also improve from using kitchen range hoods that have better capture efficiency. This latter control strategy would likely have co-benefits in lowering the indoor concentrations of other contaminants emitted from cooking, such as acetaldehyde and acrolein as describe earlier.

From the 2007 Economic Census data, there are about 16,500 food and beverage stores in California (Census, 2009) employing about 323,000 workers. Not all the rooftop units are ready to use higher efficiency air filters. But for stores where this change is possible, using higher efficiency air filters can be an effective way to lower PM indoors. Higher efficiency filters would benefit store employees and the general population who shop at grocery stores. While there are associated energy and filter replacement costs to using higher efficiency air filters, those costs are likely to be modest.

6.3 Natural Ventilation

Retail stores that rely on natural ventilation as their sole or main method of ventilation save on energy costs, as long as heating systems are not operated when doors are open. This study sampled five of such stores in north coastal California, and also one store in south coastal California. The availability to maintain comfortable indoor temperature by opening entrance doors, loading doors, etc. determines if stores can rely on natural ventilation alone. Some of the stores sampled would switch to mechanical ventilation when it is too hot or cold outside. Noise, dust, security concerns may also deter stores from using natural ventilation. This study found more outside air in stores that used natural ventilation (average = 1.5 h^{-1}) compared to mechanical ventilation (average = 1 h^{-1}). In two of the stores, ventilation rates were measured again with HVAC running. Ventilation rates in both stores were lower than when when natural ventilation was used, further confirming this finding.

However, there may be health concerns associate with higher levels of ozone and PM indoors in stores that rely on natural ventilation (Dutton, 2013). The average I/O ratio of ozone estimated from this study was 0.48 for stores that used natural ventilation, which is higher than the stores that are mechanically ventilated (average = 0.37). This is likely the combined result of higher air change rates with the outdoors, and also a lower deposition loss to indoor surfaces in naturally ventilated stores.

Table 20 shows the predicted difference in indoor ozone would be 13 ppb if the outdoor concentration was very high at 70 ppb, which is the 8-hour ambient standard in California established by the Air Resources Board. If the outdoor ozone level equals half the ambient standard at 35 ppb, the difference in indoor ozone between a naturally ventilated versus

mechanically ventilated store is about 6 ppb. The deposition loss rates shown in Table 20 are averaged from six stores that used natural ventilation (five furniture/hardware stores and one apparel store), and another six stores (four furniture/hardware stores and two apparel stores) that are mechanically ventilated. Using these parameters, the estimated I/O ratio for a naturally ventilated store is 0.53, and for a mechanically ventilated store is 0.35.

Table 20 Predicted indoor ozone levels in stores that used natural versus mechanical ventilation.

		Natural Ventilation	**Mechanical Ventilation**
ACH, k		1.5 h^{-1}	1.0 h^{-1}
Deposition loss, k_{dep}		1.4 h^{-1}	1.9 h^{-1}
	C_{out} **(ppb)**	C_{in} **(ppb)**	C_{in} **(ppb)**
Typical Outdoor Ozone	35	18	12
High Outdoor Ozone	70	37	24

Depending on the outdoor ozone level, the difference in indoor ozone between a naturally ventilated and a mechanically ventilated store may range between 6 ppb (if C_{out} = 35 ppb, see Table 20) and 13 ppb (if C_{out} = 70 ppb). Jerrett et al. (2009) estimated that the relative risk of death from respiratory causes associated with an increment in ozone concentration of 10 ppb was 1.1040 (95% confidence interval, 1.010 to 1.067). The study considered long-term ozone exposure and mortality of four exposure groups ranging from 33 to 53 ppb, 53 to 57 ppb, 57 to 63 ppb, and 63 to 104 ppb. However, whether the same relative risk from studies such as Jerrett et al. (2009) can be applied to indoor levels remains a question. For example, a recent review on the health effects of ambient ozone concluded that "[q]uestions related to thresholds and nonlinear ambient ozone-mortality relationships remain open at the present time" (McClellan, 2009). The predicted indoor ozone levels in retail stores shown in Table 20 are also lower than the outdoor levels. So even though ambient concentration-response curves exist and are considered in the determination of ambient standards, they are not clearly suitable for use here because of the extrapolation that is required to the low levels predicted for indoors.

In the coastal regions of California where the climate is mild for natural ventilation to be used, daytime ozone levels are more likely in the 20 to 40 ppb range. This means even smaller differences would be predicted between the ozone levels in naturally ventilated versus mechanically ventilated stores.

Besides the concern of higher ozone, higher particle levels were also measured in stores that used natural ventilation. For PM2.5, the I/O ratio from the six naturally ventilated stores was 0.91. For the six mechanically ventilated stores, the PM2.5 I/O ratio was 0.71 on average. The difference is less for PM10, I/O ratio = 0.88 for naturally ventilated stores, and 0.77 for mechanically ventilated stores. This is because a larger fraction of the particles are deposited to indoor surfaces, which is a process that occurs regardless of the ventilation mode used.

Table 21 shows the predicted indoor PM2.5 and PM10 in a naturally ventilated versus mechanically ventilated store.

Table 21 Predicted indoor PM2.5 and PM10 in stores that used natural versus mechanical ventilation.

	Outdoor		**Indoor – Natural Ventilation**		**Indoor – Mechanical Ventilation w/MERV 8 Filters**	
Typical indoor emissions: 10 μg/h-m^2 (PM2.5) and 50 μg/h-m^2 (PM10)						
	PM2.5 (μg/m^3)	**PM10 (μg/m^3)**	**PM2.5 (μg/m^3)**	**PM10 (μg/m^3)**	**PM2.5 (μg/m^3)**	**PM10 (μg/m^3)**
Typical Outdoor PM	10	20	10	20	7	16
High Outdoor PM	35	50	33	41	20	27

The parameters used to predict the indoor PM concentrations are the same as used previously: air change rate of 1.5 h^{-1} for natural ventilation, 1 h^{-1} for mechanical ventilation (0.9 h^{-1} from outside air intake, 0.1 h^{-1} from infiltration, and 2.9 h^{-1} of recirculation). Naturally ventilated stores have indoor PM levels that are close to the outdoor levels. These results apply only to stores that do not have indoor emissions. In this example, the mechanically ventilated store is modeled to use MERV 8 air filters. The benefits of mechanical ventilation in maintaining lower PM levels in retail stores are more pronounced when the outdoor PM levels are high, as shown in Table 21.

There is convincing evidence from epidemiological studies that found that increased PM2.5 is associated with several acute outcomes and chronic mortality (Pope, 2002, Laden, 2006). Each 10 μg/m^3 increase in PM2.5 was associated with 4% increased risk of all-cause mortality, 6% increased risk of cardiopulmonary mortality, and 8% in lung cancer mortality (Pope, 2002). A follow-up study of the Harvard Six Cities adult cohort study found even higher mortality risk ratios associated with each 10 μg/m^3 increase in PM2.5 (Laden, 2006). Based on the evidence from these studies, there may be significant health concerns with stores that rely on natural ventilation. This is because retail stores typically are located near busy roadways for easy access. Vehicle traffic is often heavy from visiting customers as well as truck deliveries of merchandise. The result is that the outdoor PM levels tend to be elevated near retail stores. From the simple calculations shown in Table 21, particle removal using an air filter of MERV 8 or greater can lower indoor PM2.5 levels by more than 10 μg/m^3 if the outdoor levels are high, such as at 35 μg/m^3 which is the California 24-hour ambient standard. If natural ventilation were to be used, retail stores would benefit from having the option to use mechanical ventilation when the outdoor air quality is poor.

7 Conclusions

This study measured ventilation rates and contaminant concentrations at nineteen retail stores in California, including seven grocery stores, seven furniture/hardware stores, and five apparel stores. Data show that all except one store had ventilation rates exceeded the minimum requirement of Title 24. For stores that chose to rely on natural ventilation, air exchange rates tended to be higher than in stores that were mechanically ventilated.

Formaldehyde, acetaldehyde, and acrolein concentrations measured in some stores exceeded the most stringent chronic health standards. Concentrations of other VOCs were substantially lower than health guidelines. The most commonly found VOCs in retail stores include hexanal, d-

limonene, and D5-siloxane, and nonanal. The sampling methods for measuring VOCs using sorbent tubes and aldehyde cartridges are well established. Acrolein concentrations reported from this study used a pentafluorophenyl hydrazine (PFPH) sampling method that is still under development. Data collected from this study points to cooking as an important source of acrolein emissions in grocery stores.

Retail stores have varying levels of indoor PM relative to the outdoors. Measurements of PM mass using filter-based method and PM counts using real-time monitors all show that grocery stores had significant PM sources, most likely because of cooking. In all stores, particles in the smallest size bin, i.e. 0.3 to 0.5 μm had the highest count concentrations. Analyses of PM mass and count concentrations further suggests PM <0.3 μm contributes a large fraction of the total, especially inside grocery stores.

All the stores maintained low level of CO_2 concentrations however this may be because field sampling was conducted during off-peak hours (week days, and excluding holiday seasons). ASHRAE 62.1 corresponds approximately to a recommendation that the CO_2 concentration indoors is no greater than 700 ppm above the outdoor air levels. This condition was met in all the stores by a large margin on the day of the sampling. Very low concentrations of CO were measured both outdoors and indoors.

The average outdoor ozone concentrations measured outside of the stores varied greatly between 10 and 70 ppb. The observed indoor-outdoor ratio was used to estimate the surface deposition rate. There are differences in the surface removal rate by store type, with higher removal rates are estimated in apparel stores (average = 2.7 h^{-1}), and lower rates estimate in furniture/hardware stores (average = 1.4 h^{-1}). Grocery stores have an average ozone deposition loss rate k_d = 1.8 h^{-1}. This general trend, though uncertain because of the small sample size, agrees with observations that there are more soft materials in apparel stores that would remove ozone more effectively than hard surfaces in hardware stores. Stores that used natural ventilation during sampling also have k_d that is lower (average = 1.4 h^{-1}) than stores that used mechanical ventilation (average = 1.9 h^{-1}).

Source strengths of VOCs, PM2.5, and PM10 were computed and normalized to the store floor area. Hexanal and acetone are the two VOCs with the highest emission rates found, followed by D5-siloxane, acetaldehyde, d-limonene, and formaldehyde. The emission rate estimates agree well with estimates from two other field studies that were recently conducted on retail stores: (i) small and medium commercial buildings (SMCB) in California, and (ii) retail stores in Texas and Pennsylvania (ASHRAE project). Certain characteristics emerged that differ by store types. For example, the source strengths of acetaldehyde and acrolein in grocery stores are much higher than in the other stores. Source strengths of formaldehyde tend to be the highest among furniture/hardware stores. Acrolein was not measured by the other two field studies on retail stores. Sampling from five grocery stores with cooking as the likely source suggests an acrolein emission rate of about 45 $\mu g/h\text{-}m^2$. In other stores without cooking, the average emission rate was about 8 $\mu g/h\text{-}m^2$.

Estimates of PM2.5 and PM10 emission rates took into the account filter efficiency, surface deposition, and the effect of infiltration and recirculation on indoor levels. Because many

assumptions were necessary, the resulting estimates have a high uncertainty. However, using these estimates, cooking in grocery stores contributing to about 70% of the indoor PM2.5 measured. This is similar to estimates by the ASHRAE project from measurements of two grocery stores. In this study, furniture/hardware and apparel stores did not have significant indoor PM sources. Estimate of indoor contributions to indoor PM was about 30% in stores that kept their entrance door closed, and 10% in stores that rely on natural ventilation with entrance door left open.

With respect to Title 24, this study made three observations that are important to setting health-protective ventilation requirements. First is that even though formaldehyde levels remain high relative to health guidelines, measurements suggest that source strengths might be decreasing. Comparisons with SMCB and the ASHRAE project suggest that adoption of the California ARB's Composite Wood Products Airborne Toxic Control Measure on formaldehyde emissions may help to mitigate this potential concern in retail stores. There is further evidence that using low emitting materials is an effective strategy to archive low formaldehyde levels even in recently renovated retail stores.

Secondly, the analysis of source strength suggests cooking as a major source of indoor contaminants in grocery stores. While it is beyond the scope of this work to evaluate the cost and efficiency of control technologies, there are options other than increasing the store ventilation rate that may be more suited to address this problem. For example, better capturing by exhaust hoods of the cooking fumes containing acrolein, acetaldehyde, and particles can improve indoor air quality. Using higher efficiency filters, such as MERV 13 instead of MERV 8, will also lower indoor particle levels.

Thirdly, natural ventilation in retail stores brought in sufficient outside air provided that the entrance door is left open. As a result, indoor ozone levels tend to be higher in naturally ventilated stores. More importantly, the higher indoor particle concentrations in naturally ventilated stores, at levels that approach the outdoor levels may have significant adverse implications for health. Retail stores that are situated in areas with heavy traffic and poor ambient air quality may benefit from using mechanical ventilation instead of natural ventilation.

In conclusion, this study found that retail stores in California are generally ventilated at rates above the minimum requirement of Title 24. And yet, key contaminants, such as formaldehyde, acrolein, and particles, still show concentrations that exceed health guidelines. For this reason, retail stores should not lower their ventilation rates without first reducing indoor emissions of these contaminants. It is recommended that source reduction and filtration be considered as energy efficient ways to improve indoor air quality in retail stores.

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