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Contaminant levels and source strengths in U.S. retail stores–A pilot study

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SUMMARY
Retail stores have many sources of indoor air contaminants that can cause potential health and odor concerns. One way to control exposure to these contaminants is to provide adequate ventilation. This study aims to characterize the whole-building emission rates of contaminants in certain retail types. The pilot study included two grocery stores and three furniture stores in northern California. We measured simultaneously the building ventilation rates by SF_6 decay and contaminant concentrations in each store for one to two days. Contaminants were measured at multiple indoor locations and at one outdoor location near the building. Formaldehyde, acetaldehyde, and acrolein are three compounds with concentrations above health guidelines in some stores. In several cases, indoor concentrations of certain VOCs and PM were higher indoors than outdoors, suggesting potential indoor sources. Our goal is to characterize the range of contaminant source strengths in 25 to 30 stores in California.

KEYWORDS
Building ventilation, air-exchange rate, VOCs, particles, indoor-outdoor ratio

1 INTRODUCTION

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 sufficient or excessive for controlling contaminant levels. Studies have found the types of contaminants and their concentrations to vary considerably among different retail stores (Loh et al. 2006; Bennett et al. 2011); thus, the required VRs may also vary among types of stores. For example, Loh et al. (2006) found that furniture stores tend to have higher formaldehyde concentrations than other retail types. Bennett et al. (2011) also found statistically significant differences by building types for formaldehyde and a number of other VOCs, such as ethylbenzene, m/p-xylene, o-xylene, chloroform, PCE, naphthalene, etc.

The goal of this study is to identify contaminants in retail stores that should be controlled via ventilation, and to determine the minimum VRs that would satisfy the health and odor criteria. This paper summarizes contaminant measurements and ventilation rates in five California stores, which is a subset of the 25 to 30 stores that are being planned. In future work, this data will be used to compute the whole-building source strengths. Our focus is on VOCs because there are indoor sources, such as from building materials and merchandise. We focus on several store types: grocery, home/furniture/hardware, and clothing, from which we can see if source strengths of contaminants vary among them. Findings of this work will inform future VR standards in Title 24.

2 MATERIALS/METHODS

Five retail stores located in California's Alameda and Contra Costa counties were sampled (Table 1). The two grocery stores are relatively new and are mechanically ventilated. The furniture stores are older buildings that are naturally ventilated. These stores volunteered to participate in our study. They were not selected based on criteria and so they make a convenience sample. We counted the number of occupants, including store workers and customers, a few times during the day of sampling (typically from 10 am to 5 pm). Table 1 shows the maximum number of people we observed and basic building characteristics. Major renovations had not occurred in any of the stores in the past year.

Table 1. Retail stores sampled in pilot study.

Building	Sample Day(s)	Floor Area (m^2)	Volume (m^3)	Occupants Observed	Year Built	Ventilation
Grocery Store G01	2011/9/27–28	3270	24920	260	2009	Mechanical
Grocery Store G02	2011/10/6–7	1840	12335	180	2001	Mechanical
Furniture Store F01	2011/10/24–25	640	2540	5	1920s	Natural
Furniture Store F02	2011/11/1	1530	9345	10	1920s	Natural
Furniture Store F03	2011/11/2	678	2480	8	1900s	Natural

The experimental setup in each store is similar. Indoor samples for multiple pollutants were collected from a central location on the sales floor. This is supplemented by two to three additional locations on the sales floor where only VOC samples were collected. Outdoor samples were collected near the main point of entry of outside air. For the mechanically ventilated stores, the outdoor location was near the rooftop air intake. Outdoor sampling was conducted near the store front door for the three furniture stores. Stores F01 and F02 left this door opened most of the time, but not F03. In stores G01, G02, and F02, there were other large openings in addition to the front door, such as the loading docks. The grocery stores have other indoor areas that were not included in our study space, such as offices, employee breakroom, and warehouse. In addition to the sampling of VOCs, O_3, and PM that are described in this paper, CO, CO_2, temperature, and relative humidity were also monitored in real time.

Ventilation Rate

SF_6 was released in the store in the early afternoon. Three Miran SapphIRe® Model 250B infrared gas analyzers were used to monitor the tracer concentrations in real time at three locations in the store. Different amount of tracer gas was released on the sales floor depending on the store size to achieve an initial concentration of about 1,000 ppm. A large fan was used in the naturally ventilated stores to help with mixing for approximately 20 minutes post-release. Air-exchange rate was computed from curve fitting to the exponential decay in SF_6 concentrations.

Volatile Organic Compounds

VOCs were collected using multi-bed Carbopack X® and Carbopack B® sorbent tubes. One-hour samples were collected at 0.1 Lpm. Sample flow was monitored using flow sensors, and were verified using a primary air flow calibrator (Gilibrator®). Forty-four target VOCs were qualitatively and quantitatively analyzed by thermal desorption-gas chromatography/mass spectrometry generally following U.S. EPA Methods TO-1 and TO-17. Multi-point internal standard calibrations were performed using pure compounds and 1-bromo-3-fluorobenzene as the reference compound. A duplicate set of samples was collected at all times. 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 samples.

Volatile carbonyl samples were collected using dinitrophenyl hydrazine (DNPH)-coated cartridges (Waters Sep-Pak®). Ambient ozone was removed with potassium iodide scrubbers preceding each DNPH sampler. 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 HPLC with UV detection at λ_{max} = 360 nm (Agilent 1200). A calibration curve for quantification was carried out using authentic standards of the formaldehyde-DNPH hydrazone.

In addition, we collected acrolein samples using pentafluorophenyl hydrazine (PFPH)-coated cartridges that were prepared in our laboratory. Silica cartridges were cleaned by passing 2 mL of acetonitrile solution and then air-dried. 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. We are still in the process of refining this recently-developed acrolein method for field sampling, therefore the results presented in this paper are preliminary. We prepared acrolein standards to obtain calibration curves prior to each analysis. The limit of detection was 0.2 ng/m^3 (~0.1 ppb) for stores G01 and G02, where one-hour samples were collected at 0.1 Lpm. At stores F01, F02, and F03, sampling duration was increased to two hours, so the limit of detection was approximately 0.05 ppb. Breakthrough was estimated to occur at ~0.02 mg, or sampling at 100 ppb for 15 hours.

Ozone

Concentrations of O_3 were monitored using real-time gas analyzers (2BTech® Model 205) at the central indoor location and also outdoor. The gas analyzers were calibrated by the manufacturer. We checked that the indoor and outdoor units agreed with one another before and after sampling. Limit of detection is 2 ppb. Due to operator error, O_3 data was lost from stores G01 and F03. In stores G02, F01, and F02, only one day of data was recorded.

Particulate Matter

Particle counts and mass concentrations were measured at the central indoor location and also outdoor. Real-time particle counts was monitored using MetOne® Optical Particle Counter Model BT-637 in six channels: >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. Particle mass was collected onto PTFE membrane filters, which were measured gravimetrically in a temperate and relative humidity controlled chamber. PM2.5 and PM10 samples were collected using SKC Personal Environment Monitors at 10 Lpm for approximately 6 hours. Two field blanks were collected on each sampling day. Measurement error is ±0.80 μg based on field blanks, or approximately ±0.23 μg/m^3 for a 6-hour sample.

3 RESULTS

Table 2 shows the air-exchange and ventilation rates estimated from SF_6 decay curve fitting. In most cases, results from the three Miran analyzers agreed with one another to about 10%. However, substantial differences were resulted from the three Miran analyzers in store F03, which was a relatively small store with very low ventilation. This is likely due to limited mixing in the building. The mixing fan generated a uniform SF_6 concentration in the store initially, but once the fan was turned off, SF_6 concentrations varied in different parts of the store. The estimated air-exchange rates on two days were within 10% for the two mechanically ventilated stores G01 and G02. For store F01, which was naturally ventilated, the difference between the two days was 20%. Relative to California's Title 24 standards, all stores met the minimum ventilation rate on a per person basis (7 L/s-person), but store F03 did not meet the per floor area requirement (1 L/s-m^2).

Table 2. Air-exchange and ventilation rates estimated from SF_6 decay.

Building	Air-Exchange Rates* (h^{-1}) Day1	Day2	Ventilation Rates ($L/s\text{-}m^2$)	(L/s-person)
Grocery Store G01	0.6 (0.6–0.7)	0.7 (0.7–0.8)	1.4	18
Grocery Store G02	2.0 (1.8–2.1)	1.8 (1.7–1.8)	3.5	36
Furniture Store F01	1.3 (1.2–1.3)	1.0 (0.98–1.0)	1.3	160
Furniture Store F02	2.8 (2.6–3.0)	--	4.8	740
Furniture Store F03	0.4 (0.2–0.6)	--	0.4	30

*Average and range, as shown in parenthesis, of results estimated from three Miran analzyers.

Table 3 compares the outdoor and indoor concentrations of formaldehyde, acetaldehyde, and acrolein measured in the five stores. Indoor concentrations measured at different locations were averaged, and the concentration ranges (minimum to maximum) are also shown. Higher concentrations were found indoors. Grocery stores G01 and G02 had lower formaldehyde, but higher acetaldehyde and acrolein concentrations, than the furniture stores. The day-to-day and spatial variability for these three chemicals is relatively small compared to the between-store differences. The indoor concentrations in some stores are higher than the chronic health guidelines for these three compounds: 7.3 ppb for formaldehyde (OEHHA risk-based reference exposure levels), and 5.0 ppb for acetaldehyde and 0.009 ppb for acrolein (U.S. EPA IRIS reference inhalation concentrations). A recent California study of 40 small and medium commercial buildings found formaldehyde and acetaldehyde concentrations of about 10 ppb in grocery stores and restaurants (Bennett et al. 2011). Among the 40 buildings sampled were 7 retail stores of various types, where the concentrations of formaldehyde and acetaldehyde were about 20 ppb and 3 ppb, respectively. These levels were similar to Table 3. To the best of our knowledge, acrolein concentrations have not been previously measured in commercial buildings.

Table 3. Outdoor and indoor volatile carbonyl concentrations measured in five stores.

Building	Formaldehyde (ppb)		Acetaldehyde (ppb)		Acrolein (ppb)	
	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor
G01–Day 1	2.4	7.8 (7.5–8.5)	1.6	16.0 (14.3–19.3)	6.5	17.2 (16.2–18.7)
Day 2	6.6	8.4 (8.2–8.8)	4.2	14.5 (13.8–15.8)	4.6	15.5 (14.1–16.9)
G02–Day 1	1.4	4.2 (3.8–4.9)	1.0	8.6 (7.8–9.8)	3.2	2.7 (1.8–4.2)
Day 2	2.7	4.3 (4.1–4.6)	1.3	11.5 (8.9–14.8)	0.2	2.2 (0.8–2.8)
F01–Day 1	1.1	17.3 (14.7–21.2)	1.1	2.5 (2.2–2.7)	0.5	1.5 (1.2–1.7)
Day 2	1.7	19.5 (13.9–22.8)	1.2	3.3 (1.5–4.9)	0.6	1.0 (0.2–2.0)
F02	3.3	13.2 (11–15.7)	1.4	2.4 (1.8–2.9)	0.07	0.6 (0.05–1.1)
F03	1.9	20.8(14.5–26.8)	1.5	4.8 (4.6–5.2)	0.03	0.2 (0.02–0.3)

VOCs sampled by sorbent tubes were low (~1 ppb or less). The most commonly measured compounds with indoor concentration >1 ppb are hexanal, d-limonene, α-pinene, and toluene. The small and medium commercial buildings study also found VOC concentrations, other than the three listed in Table 3, to be low. Bennett et al. (2011) found only six of the 27 VOCs with mean concentrations >1 ppb in 40 buildings sampled: 2-butoxyethanol, d-limonene, D5-siloxane, toluene, decanal, and hexanal. More detailed analysis of VOC results is underway.

Figure 1 shows the outdoor and indoor O_3 concentrations at the three stores where we collected data. Store G02 had a similar indoor-outdoor ratio of O_3 as in store F01, despite that it had a higher air exchange with the outdoors. This may be because store G02 is mechanically ventilated, so there were losses of O_3 as the outside air passes through filters (Zhao et al. 2007). Store F02 had higher air exchange with the outdoors than store F01, this is reflected in

a higher indoor-outdoor ratio of O_3 compare to store F01. The data presented here can be used to estimate parameters such as the deposition rate to indoor surfaces (Weschler 2000).

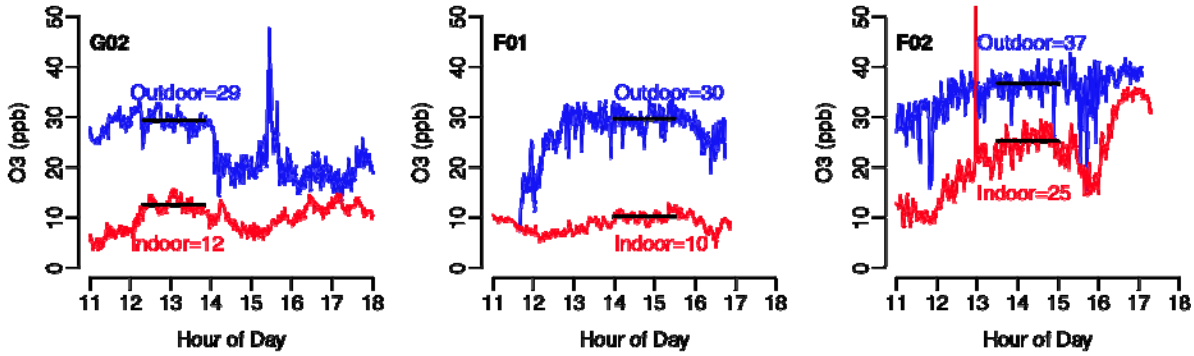

Figure 1. Outdoor and indoor concentrations of O_3 measured in three stores. Mean concentrations measured over a period of 90 minutes were indicated.

Particle mass concentrations measured gravimetrically are shown in Table 4. Stores that are naturally ventilated (F01, F02, and F03) all had indoor-outdoor (IO) ratio of about one. There is a substantial difference in the IO ratios of the mechanically ventilated stores. Overall, G01 had fewer particles indoors than outdoors, and the reverse is true for G02. We observed that G01 had MERV 15 air filters, which are more effective than the MERV 8 air filters used in G02. There were also possible indoor sources of particles in G02. Figure 2 shows the particle number distributions measured by the optical particle counter in two stores. On Day 1 in G02, the particle counts with diameter <1 μm were higher indoors than outdoors.

Table 4. Particle mass concentrations of PM2.5 and PM10 measured outdoors and indoors.

	Outdoor			Indoor			IO Ratio	
Building	Sampling Time	PM2.5 ($\mu g/m^3$)	PM10 ($\mu g/m^3$)	Sampling Time	PM2.5 ($\mu g/m^3$)	PM10 ($\mu g/m^3$)	PM 2.5	PM 10
G01–Day 1	10:27–16:53	11.4	21.9	10:37–17:02	8.2	10.8	0.7	0.5
Day 2	09:06–16:08	20.9	40.4	08:25–16:01	6.8	12.5	0.3	0.3
G02–Day 1	09:43–14:04	4.4	25.1	09:29–16:00	10.9	18.7	2.5	0.7
Day 2	08:56–16:04	4.3	8.1	08:13–16:28	12.5	19.2	2.9	2.4
F01–Day 1	12:35–17:23	13.0	25.0	12:02–17:35	12.8	19.8	1.0	0.8
Day 2	11:44–16:36	13.0	29.9	11:34–16:47	14.3	22.0	1.1	0.7
F02	10:54–17:00	7.2	19.2	10:56–17:16	6.4	20.1	0.9	1.0
F03	10:33–17:02	6.2	21.8	10:37–17:06	6.4	16.7	1.0	0.8

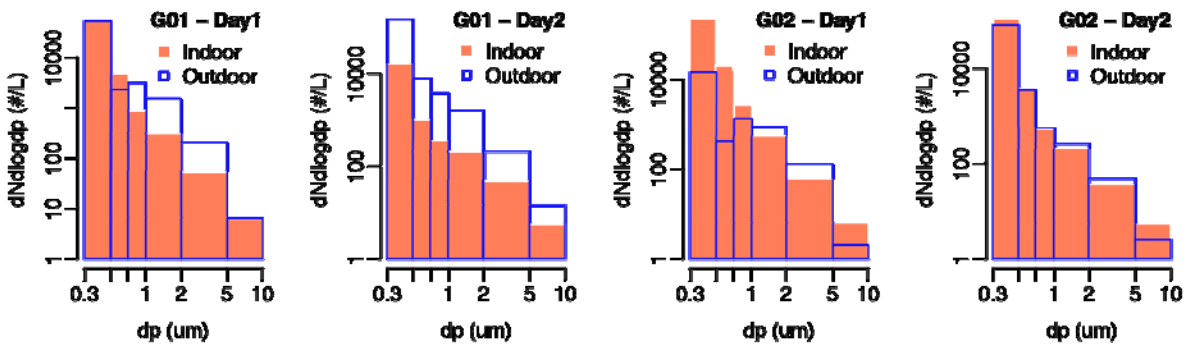

Figure 2. Particle number distributions measured in two stores. Distributions were computed from the daily averaged particle counts measured during the sampling periods (see Table 4).

4 DISCUSSION

We plan to compute the whole-building source strength for contaminants that have sufficient indoor sources based on the data presented here. Results from the five stores suggest that formaldehyde, acetaldehyde, and acrolein are likely the contaminants that may be of health concerns. Concentrations of other VOCs were generally low (~1 ppb or less). The between-store type difference is evident from this preliminary data, which suggest ventilation needs may differ also. Our study design of collecting VOCs from a few indoor locations in stores on a given day is expected to generate a reliable dataset because contaminant concentrations did not appear to vary spatially or from day-to-day in a significant way. The SF_6 decay method also worked well and reasonable air-exchange rates were estimated. The real-time O_3 and PM data are useful for understanding the indoor and outdoor relationship in retail stores. Even though O_3 and PM tend not to have major indoor sources, they are important to consider because they can impact VOC concentrations through chemical reactions or phase partitioning. Beyond the exploratory analyses of O_3 and PM presented here, we plan to use the data to estimate parameters needed for modeling their dynamic behaviors in retail stores.

5 CONCLUSIONS

Building ventilation rates and contaminant concentrations were measured in five retail stores in California. Most stores exceeded the minimum ventilation rate of 1 L/s-m^2. VOC concentrations were generally low (~1 ppb or less), but a few compounds had measured concentrations above health guidelines. If the current ventilation rate standards have the goal of protecting health, identification of the relevant indoor contaminants and knowledge of their source strengths is needed. The data presented here, together with additional stores to be sampled in California, will form a dataset providing the needed information for determining the minimum ventilation rate requirements.

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