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Interpreting time-resolved residential monitoring data to characterize emissions of volatile organic compounds from occupant activities

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SUMMARY
Occupant associated emissions of volatile organic compounds (VOCs) can materially influence indoor air quality (IAQ). Knowledge of emission factors can contribute to improved exposure and risk assessments and also can aid in designing effective control measures to improve IAQ. The advent of instruments capable of sensitive, speciated VOC measurements with high time resolution allows for characterizing occupant-associated VOC emissions from observational monitoring studies conducted in real buildings under normal occupancy. This presentation highlights VOC emissions associated with residential activities, such as cooking and cleaning.

KEYWORDS
Indoor VOCs; VOC sources; emission rates; cooking; cleaning

1 INTRODUCTION
Dating back to the classic works of von Pettenkofer (ca. 1870) and Yaglou (ca. 1930), emissions from occupants have been recognized as an important source of indoor air pollution. Olfactory perception of occupant-associated bioeffluents has served as the primary basis for ventilation-rate guidelines and standards. In recent decades, occupant activities indoors, such as cooking and cleaning, have become recognized as important contributors to the emissions of volatile organic compounds (VOCs). Source characterization studies using analytical instruments have most often been conducted in controlled facilities, such as environmental chambers and research houses (e.g., Singer et al., 2011; Liu et al., 2017). Recent advances in scientific instruments for measuring organic composition of air are enabling improved emissions characterization for all indoor sources, including those from occupants and their activities. One such instrument is the proton-transfer-reaction time-of-flight mass spectrometer (PTR-ToF-MS), which can measure hundreds of compounds simultaneously, with high time resolution (sampling time of seconds), exquisite sensitivity (down to 10 ppt or better), and very good species discrimination. In this paper, we will describe the utilization of PTR-ToF-MS to assess occupant-associated emission factors from a residential field monitoring campaign, focusing on emissions from the occupant activities.

2 METHODS
Emissions are usefully characterized in terms of chemical species emitted per some useful quantification unit, such as per unit activity. With time-resolved measured concentrations, emissions can be quantified using the principle of material balance or mass conservation.

As a broad classification that is useful for many VOCs, emission sources are sorted into contributions from outdoor air, contributions from non-occupant associated indoor sources (such as building materials and furnishings), and occupancy-associated contributions. The occupant-

associated emissions can be further subdivided into those that arise directly from the occupant and those that are from the occupant's activities. Those emissions directly from the occupants are further subdivided into endogenous emissions, such as in exhaled breath reflecting human metabolism, and exogenous emissions, such as from personal care products applied to the body. We have found that measurements in classroom settings or other densely occupied spaces with restricted activity can be effective for characterizing emissions directly from the occupants (Tang et al., 2016). Measurements in residences where occupancy rates are lower have been particularly useful for characterizing emissions associated with occupant activities, such as cooking and cleaning. In both types of environments, it has been important to characterize air-change rates as a major removal mechanism in the material balance. It has also been important to measure outdoor concentrations to assess their contributions to indoor levels. And, it has been important to acquire time-resolved metadata such as activity type and intensity. In carrying out analyses, we have applied the integral form of material balance equations to quantify emissions over finite intervals, such as the duration of meal preparation.

3 RESULTS AND DISCUSSION

Our first residential field study entailed monitoring in an ordinarily occupied single-family dwelling in Oakland, California. (See also abstract by Yingjun Liu et al.) Sampling was undertaken continuously for an 8-week period in summer 2016 and a 5-week period in winter 2017. The PTR-ToF-MS was configured to sample twice per hour for five-minute periods from each of five locations inside plus one location outside the residence. An array of more than 50 wireless sensors was used to monitor the status of elements that could influence indoor air composition. Constant tracer-gas release was employed to characterize air-change rates. Occupants maintained presence/absence logs and also noted the time and nature of cooking and cleaning events. The time series of many VOCs exhibited clear episodic short-term enhancements, which we could attribute to specific identified occupant activities. Emission strengths of specific VOCs are quantified on a per-event basis; a few examples follow. During breakfast, emissions on the order of ~1 mg of pyridine and ~ 40 mg of ethanol occurred, in association with making coffee and toasting bread, respectively. When baking using the oven, we found that tens of VOCs were emitted, such as ethanol (~ 5 g/event), acetaldehyde (~ 100 mg/event), pentanal (~ 5 mg/event) and methanethiol (~ 0.5 mg/event). When cleaning hard surfaces using a popular brand of window cleaner, we determined emissions to be ~50 mg of acetone and a few mg of 2-hexoxyethanol per event. We could detect and quantify emissions of the disinfectant chloramine (0.1-1.0 mg/event) in association with various uses of tap water, such as showering.

4 CONCLUSIONS

Occupants pollute indoor air. Improved knowledge about emissions from occupants and their activities can serve as the foundation for better exposure assessments and more efficacious engineering controls. Advanced instrumentation that enables sensitive real-time monitoring of speciated VOCs enables studies to characterize emissions in normally occupied indoor environments. Such real-world assessments can complement studies undertaken in test chambers.

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