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Spatially and temporally resolved emissions of volatile organic compounds in a residence

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SUMMARY
The present study provides detailed investigation of source characteristics and emission dynamics of volatile organic compounds (VOCs) in a single-family house in California, utilizing time- and space-resolved VOC measurements for eight weeks in summer and five in winter. In total, about 300 VOCs were measured. Spatially resolved VOC measurements, in conjunction with airflow investigation using multi-tracer technique, reveal that VOCs in the living space were mainly emitted directly into the living space, with minor contributions from coupled interior spaces (crawlspace and attic) and from outdoors. Time series of most indoor VOCs were characterized by elevated baseline levels with episodic enhancements. The high baseline level highlights the importance of continuous emissions into the house from building materials and contents. Associated emissions rates were quantified with 2-h resolution. Influences of environmental parameters on emissions were explored. Episodic enhancements were associated with occupants and their activities, such as cooking and personal-care-product use.

KEYWORDS
indoor VOCs; VOC sources; VOC emission rates; residential environment

1 INTRODUCTION
Elevated concentrations of volatile organic compounds (VOCs) is a major concern for residential indoor air quality. The conventional approach to study indoor VOCs is to collect time-integrated samples followed by offline analysis (e.g., Zhu et al., 2013); temporally resolved indoor VOC data are rare. Consequently, researchers lack deep insight regarding indoor VOC sources, emission rates, and dynamic behavior in real indoor settings. The current study carries out spatially and temporally resolved measurements of VOCs, combined with extensive auxiliary data acquisition, to characterize the sources and to explore the dynamic behavior of VOCs in a residence during normal occupancy.

2 METHODS
A single-family house in the San Francisco Bay Area, USA, was selected for the study. The house was built in the 1930s using conventional wood-frame construction. Two field campaigns were undertaken, one in the summer (non-heating; 8-week duration) and a second during winter (heating; 5-week duration). Both campaigns included vacant and occupied periods.

A proton-transfer-reaction time-of-flight mass spectrometer (PTR-ToF-MS) was used for making temporally and spatially resolved measurements of VOC composition, switching between 6 different inlet locations (outdoor, kitchen, bedroom area, basement, crawlspace, and attic) at 5-minute intervals. Other trace gases, such as CO_2 and O_3, were measured simultaneously. Three deuterated alkene tracers were continuously released at constant rates from different locations in

the house, to determine dynamic air-exchange and interzonal transport rates. An extensive set of metadata was collected, including the utilization of more than 50 wireless sensors to monitor time-resolved room occupancy, appliance use, door/window open status, temperature, and humidity.

3 RESULTS AND DISCUSSION

Altogether, more than 600 mass peaks, corresponding to about 300 VOC species, were detected throughout the two sampling campaigns. For 80% of observed ions, the mean signals in the living space (kitchen and landing) were more than double the outdoor levels. For one third of the ions, levels in the living space were at least 10 times higher than outdoors. As indicated by tracer gases, air from the crawlspace was transported upward into the living space and the attic. Conversely, and somewhat surprisingly, transport from the attic to the living space was almost undetectable (< 1%). The crawlspace air composition was similar to that outdoors. Consequently, VOC transport from the crawlspace and attic (coupled spaces) were both minor sources to the living space for this house. A key conclusion is that VOCs observed in the living space were mainly emitted from sources in the living space itself. These emissions must originate from a combination of the building structure surrounding that space, the indoor contents, the occupants, and their activities.

The time series of most VOCs observed in the living space were characterized by clear episodic short-term enhancements on top of consistently elevated baseline levels. The enhancements generally occurred during human occupancy, and generally were associated with specific identified activities such as cooking and personal care product use. However, except for a few species closely tied to human activities (e.g., ethanol and cyclic siloxanes), the contribution of the episodic enhancements represented a small contribution to the mean concentrations of the indoor VOCs which would have been observed through typical time integrated sampling. Instead, the high baseline level for the majority of observed ions was a more general, and, to some extent, unexpected feature of the time series, highlighting the importance of continuous emissions from building materials and contents.

One major continuous VOC source was identified to be decomposition and emission of wood building materials. For many abundant indoor VOCs such as acetic acid and furfural, their concentrations in the unoccupied attic (without furniture) were higher than in the living space and increased strongly with temperature, suggesting emissions from wood building materials common for the two spaces. Moreover, the observed VOC composition and temperature dependence of emission rates was consistent with chemical degradation of hemicellulose (Esteves and Pereira, 2009).

4 CONCLUSIONS

The current work demonstrates that temporal patterns of VOC emissions from occupant activity are distinct from those originating from building materials and contents, and can be quantified separately and attributed to specific source categories. Decomposition and emission of wood building materials is suggested to be an important VOC source in wood-frame houses.

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5 REFERENCES

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