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Evaluation of three common green building materials for ozone removal, and primary and secondary emissions of aldehydes

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HIGHLIGHTS

- Green materials are increasingly used in modern buildings and retrofits.
- Three large-area green materials were tested with and without ozone present.
- Substantial differences in ozone removal, primary and secondary emissions occurred.
- Relative humidity did not affect ozone removal and had mixed effects on emissions.
- Large and small chamber results were generally consistent for test materials.

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ABSTRACT

Ozone reactions that occur on material surfaces can lead to elevated concentrations of oxidized products in the occupied space of buildings. However, there is little information on the impact of materials at full scale, especially for green building materials. Experiments were completed in a 68 m^3 climate-controlled test chamber with three certified green building materials that can cover large areas in buildings: (1) recycled carpet, (2) perlite-based ceiling tile and (3) low-VOC paint and primer on recycled drywall. Ozone deposition velocity and primary and secondary emission rates of C_1 to C_{10} saturated carbonyls were determined for two chamber mixing conditions and three values of relative humidity. A direct comparison was made between ozone deposition velocities and carbonyl yields observed for the same materials analyzed in small (10 L) chambers. Total primary carbonyl emission rates from carpet, ceiling tile and painted drywall ranged from 27 to 120 μg m^{-2} h^{-1}, 13 to 40 μg m^{-2} h^{-1}, 3.9 to 42 μg m^{-2} h^{-1}, respectively. Ozone deposition velocity to these three materials averaged 6.1 m h^{-1}, 2.3 m h^{-1} and 0.32 m h^{-1}, respectively. Total secondary carbonyl emissions from these materials ranged from 70 to 276 μg m^{-2} h^{-1}, 0 to 12 μg m^{-2} h^{-1}, and 0 to 30 μg m^{-2} h^{-1}, respectively. Carbonyl emissions were determined with a transient approximation, and were found to be in general agreement with those found in the literature. These results suggest that care should be taken when selecting green building materials due to potentially large differences in primary and secondary emissions.

1. Introduction

There are many sources of indoor pollution, including primary emissions, e.g., those that occur from a building material as manufactured and not from transformations (physical, biological or chemical) that occur once the material is placed in the building. However, there is also a recognition that chemical reactions between oxidants and many materials lead to secondary emissions that are important and deserving of more attention (e.g., Wolkoff, 1999; Weschler, 2004). Ozone is often the driver of interfacial chemistry in buildings, and numerous researchers have characterized ozone removal to a wide range of building materials (e.g., Morrison et al., 1998; Klenø et al., 2001; Grøntoft and Raychaudhuri, 2004; Bekö et al., 2006; Poppendieck et al., 2007; Hoang et al., 2009).

Importantly, ozone reactions with building materials result in secondary emissions of aliphatic aldehydes and other reaction products (e.g., Weschler et al., 1992; Reiss et al., 1995b Morrison and Nazaroff, 2002). Summertime indoor concentrations of ozone reaction products have been observed to be significantly greater than

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wintertime concentrations, a result of higher ozone concentrations during the summer months (Reiss et al., 1995b). Four homes in Rolla, Missouri, had substantial secondary emissions from walls, carpets, floors, and countertops in the presence of elevated ozone concentrations (Wang and Morrison, 2006). Typical secondary emissions noted in these studies include C_1 (formaldehyde), C_2 (acetaldehyde), and C_5–C_{10} (pentanal–decanal) aliphatic aldehydes. There is potential for adverse health impacts by secondary emissions of compounds like formaldehyde, a carcinogen and sensory irritant (World Health Organization, 2010), and occupant discomfort from secondary emissions of irritating and/or odorous compounds such as nonanal (C_9) (Wolkoff, 1999; Knudsen et al., 2003; Peng et al., 2009). It is possible that these secondary emissions are responsible for the health effects of what has historically been correlated with outdoor ozone. Apte et al. (2008) observed that ambient ozone correlated with indoor concentrations of some aldehydes in Building Assessment and Survey Evaluation (BASE) office buildings and concluded that ozone-initiated indoor chemistry may play an important role in the reported health of building occupants. For these reasons, efforts are being made to incorporate indoor ozone chemistry into population wide assessments of ozone and ozone reaction product exposure (Weschler, 2006; Chen et al., 2012) and to further characterize and assess the impact of short-term exposure to ozone-initiated products in indoor spaces (Wolkoff et al., 2013).

The primary objective of this research was to complete room-scale chamber experiments to determine ozone deposition velocities and reaction product formation rates for three certified green materials under varying environmental conditions. Here, we considered emissions of C_1–C_{10} *n*-aldehydes, acetone, benzaldehyde and tolualdehyde reaction products, with specific emission rates reported for major constituents and "total" carbonyls reported as the sum of the aforementioned constituents. A secondary objective was to compare these large scale results against small-scale chamber experiments reported for the same test materials (Lamble et al., 2011; Cros et al., 2012). The materials selected for study were three popular green building materials that are increasingly used in homes, commercial buildings, and schools: recycled carpet tile, perlite based ceiling tile, and recycled drywall coated with low-VOC primer and low-VOC paint. Determining ozone deposition velocity and byproduct formation rates at varying mixing and relative humidity conditions will provide important data regarding material-ozone behavior over a diverse set of indoor conditions. Comparisons between small chambers and environments representative of full-scale installations further the ability to determine if extrapolation from studies conducted at small scale is appropriate. Because of the variety of green building materials available, such efforts are warranted to promote and facilitate the inclusion of *in-situ* chemistry in testing protocols from green building materials.

2. Methodology

2.1. Building materials tested

Materials were selected based on: (1) representation of materials that cover large surface areas (walls, floors, and ceiling), (2) representation of different compositions between materials, (3) anticipation of large ranges in reactivity with ozone and reaction product yields, and (4) recent studies using small test chambers and the same materials (Lamble et al., 2011; Cros et al., 2012). In all cases, comparisons with these studies were made with only results reported for new (unused) materials. Materials selected for this research had qualities considered "green" and were certified as such by at least one organization. The carpet and drywall considered in this study had high recycled content while the perlite-based ceiling tile and architectural coatings (applied to the recycled drywall) were considered likely low-emitting and certified low-emitting, respectively.

The first material tested was recycled carpet tile (Interface FLOR), certified as green by the Carpet and Rug Institute's Green Label Plus program. It was installed by placing 25.4 m^2 of carpet on the floor of the chamber shown in Fig. 1. No underlayment was used, but each carpet tile had an approximate 0.33 cm thick rubber pad at its bottom edge, i.e., in direct contact with the chamber floor. The carpet tiles had dimensions of 50 cm × 50 cm × 0.7 cm and consisted of a 100% post-consumer content type-6 nylon loop pile of height 0.43 cm. The carpet had a total recycled content of 68%–71%.

The second material, 22.5 m^2 of ceiling tile (Eurostone, Chicago Metallic Corporation), is advertised as being eligible for United States Green Building Council's LEED Environmental Quality credits 4.1–4.4 for having no reportable VOCs present in the finished product. These ceiling tiles were installed on the chamber's suspended ceiling using a standard drop ceiling grid. The dimensions of the ceiling tiles were 60.9 cm × 60.9 cm × 2.22 cm. The ceiling tiles consisted of 50–70% by weight expanded perlite, 15%–30% by weight sodium silicate, and 5%–15% by weight kaolin. The ceiling tile density was 0.36 g cm^{-3}.

Recycled drywall was purchased from a local distributor in Austin, Texas. The drywall sheets (dimensions of 121.9 cm × 243.8 cm × 0.635 cm) contained recycled paper backing, which covered reclaimed gypsum (Sheetrock, synthetic from Galena: USG, Chicago, IL, USA). Three months before experiments, the sheets were painted with low-VOC, 100% acrylic primer (EcoSpec, Benjamin Moore & Co.) and two coats of low-VOC, 100% acrylic, flat-finish paint (EcoSpec, Benjamin Moore & Co), each certified by Benjamin Moore's internal "Green Promise" designation. The primer contained water, acrylic resin, a maximum of 15% titanium dioxide, and a maximum of 6% hydrous alum silicate. The paint contained a maximum of 25% titanium dioxide, 15% limestone, 5% silica and 5% diatomaceous earth. A coverage area of 14.9 m^2 of painted drywall was installed in the chamber for these tests.

2.2. Apparatus and procedure

Experiments were completed in a 68 m^3 climate-controlled stainless steel environmental chamber (Fig. 1). Laboratory air was continually introduced into the chamber at a measured air exchange rate of 1.1 ± 0.1 h^{-1} (mean ± 1 std. deviation) using a thermal dispersion airflow plenum probe and transmitter (Model Gtx116, Ebtron, Inc.). Average chamber temperature was 25.2 °C ± 1.0 °C. Prior to entering the chamber, laboratory air was

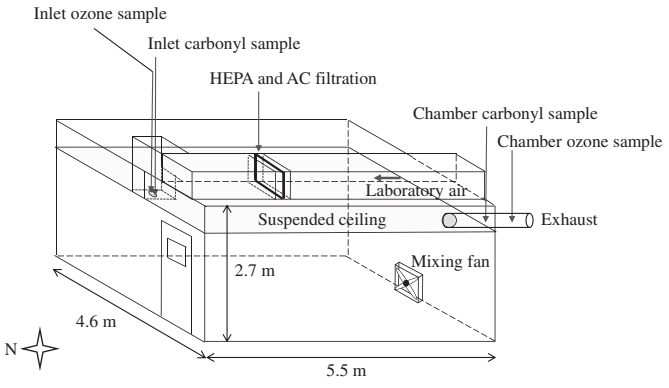

Fig. 1. Schematic of large environmental chamber for building materials testing. HEPA = high-efficiency particulate air, AC = activated carbon.