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Comparison of conventional and green building materials in respect of VOC emissions and ozone impact on secondary carbonyl emissions

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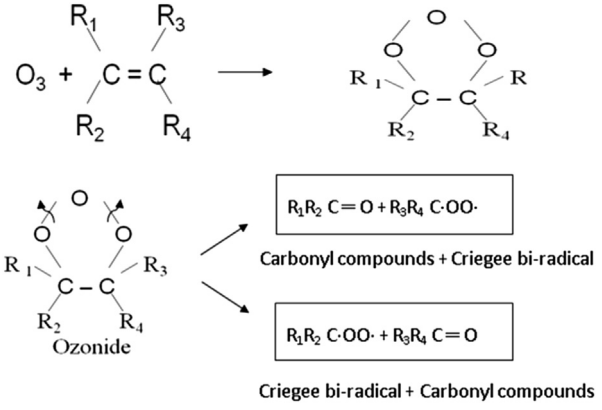
ABSTRACT

Building materials (BMs) are major contributors to indoor emission sources of volatile organic compounds (VOCs). In this study, 8 kinds of BMs (including conventional and green) for ceiling, cabinetry, and flooring commonly used indoors were tested in a 216 L chamber. Primary emissions of carbonyls (C_1 to C_8 aldehydes and ketones) at 48 h were 75–673 μg m^{-2} h^{-1} from conventional BMs, and 62–151 μg m^{-2} h^{-1} from green BMs. Primary emissions of BTEX at 48 h were 59–264 μg m^{-2} h^{-1} from conventional BMs, and 37–56 μg m^{-2} h^{-1} from green BMs. Ozone initiated molar yields of carbonyls were 0.10–2.36 from conventional BMs, and 0.13–0.86 from green BMs. Secondary emissions of carbonyls were 7–150 μg m^{-2} h^{-1} from conventional BMs, and 4–73 μg m^{-2} h^{-1} from green BMs. Green BMs had lower emissions than conventional analogs, especially for wooden flooring and gypsum board. BMs with mineral content are the most promising materials in this study, given moderately high ozone deposition velocity but generating the least byproducts. Secondary emissions determined in this study demonstrate that moderate indoor ozone concentrations may lead to increased concentrations of carbonyls, especially formaldehyde and acetaldehyde that potentially increase adverse chronic health effects.

1. Introduction

Building materials are major contributors to indoor emission sources of volatile organic compounds (VOCs). Some of VOCs are of particular concern due to their potential health impact on human [1], e.g. formaldehyde and benzene are some of the most studied pollutants since they are classified in Group 1 of human carcinogens by the International Agency for Research on Cancer [2–5]. In an INDEX project the existing knowledge worldwide has been assessed in terms of type and levels of chemicals in indoor air, as well as, the available toxicological information [6]. It was concluded that VOCs such as benzene, formaldehyde, acetaldehyde, toluene and xylenes have to be considered as priority pollutants with respect to their health effects [6].

Many studies have shown significant VOC sources indoors. For example, the main sources of aldehydes and BTEX indoors may include building materials (BMs), such as hardwood, plywood, laminate floorings, adhesives, paints and varnishes [1,7–14], adhesives and decoration materials. In addition to these primary emissions, numerous past researches also indicated that ozone reactions with BMs result in secondary emissions of aliphatic aldehydes, secondary organic aerosols and other products that are more important [15–25]. The well-established reaction mechanism is shown below.



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Weschler et al. [26] have demonstrated that with the presence of ozone (60–100 μg/m^3) in freshly carpeted stainless-steel chamber, gas-phase concentrations of VOCs including styrene, 4-VCH and 4-PCH significantly decreased while the concentrations of aldehydes (C_1–C_{10}) increased. In addition, the total concentrations of VOCs also increased markedly. Wang and Morrison [27] observed substantial secondary emissions which include C_1 (formaldehyde), C_2 (acetaldehyde) and C_5–C_{10} (pentanal-decanal) aldehydes from walls, carpet, floors and countertops in the presence of elevated ozone concentrations. Huang et al. [28] performed small-scale environmental chamber experiments on a painted hardwood panel and found that the formaldehyde concentration within the chamber is found to increase by 215.8% given an ozone concentration of 200 ppb and a reaction time of 3.0 h.

As people pay increasing attention to the impact of emissions from BMs, the market for environmentally friendly green building materials is gradually growing. Although Green Building Material (GBM) is intended to have low toxicity and minimal chemical emission, measurement of primary emissions alone may not be sufficient since secondary emissions due to ozone reactions may affect the perceived air quality in the long run [29]. Kagi et al. [30] demonstrated that natural wood material with low formaldehyde emission after being exposed to ambient ozone produced secondary pollutants, including formaldehyde, acetaldehyde, cyclohexanone and benzaldehyde.

While past studies have performed the ozone removal and primary & secondary emissions of aldehydes from building materials, this is the first study to directly compare conventional with green BMs from the region of Taiwan by pairs in terms of primary and secondary VOC emissions, including BTEX and carbonyls. The primary aim of this work is to characterize 8 kinds of BMs as indoor VOC emission sources by conducting emission measurements before and after ozone exposure in experimental chamber, including wooden flooring (WF), green wooden flooring (GWF), gypsum board (GB), green gypsum board (GGB), calcium silicate board (CSB), green calcium silicate board (GCSB), mineral fiber ceiling (MFC) and green mineral fiber ceiling (GMFC). The measurements cover mainly carbonyls, BTEX and ozone.

2. Experimental methodology

2.1. Experimental system and procedure

Fig. 1 depicts the experimental system. Laboratory experiments were conducted in a 216 L electro-polished stainless steel chamber designed under ASTM D5116-06 standard. A small fan was used to mix the air inside the chamber. The chamber system was set up at 25 °C, 50% relative humidity, and an air exchange rate of 0.5 h^{-1}. The air flow rate was 1.8 L min^{-1} with an uncertainty of 0.308 mL measured after each experiment with a bubble flow meter (Sensidyne, Model Gilibrator 2) at the outlet of the downstream chamber.

All air entering the chamber was pre-filtered through silica gel, activated carbon, and HEPA in order to get clean air. The air was pulled through a vacuum pump with a stable volumetric flow rate and conveyed through the chamber system. Uncertainty for the sampling pump flow rate is 0.308 mL. The samples of BTEX and 13 kinds of carbonyls (CARB Method 1004 DNPH Mix2) were taken intermittently for the first 96 h. During the 96 h, ozone air (i.e., air mixed with ozone) generated by an ozone generator (2B Technologies, Model 306) at a concentration of 75 ppb was then pumped into the chamber at 48 h for 4 h to observe the impact of ozone on carbonyl emissions. After 4 h of ozone exposure, ozone generator was turned off when BTEX and carbonyls were sampled and ozone inside the chamber started to decay. Ozone concentrations inlet and outlet of the chamber were continuously monitored and recorded by UV ozone analyzers (2B Technologies, Model 202) with sampling time intervals of 10 s. A schematic diagram for a standard chamber experiment to illustrate when samples were taken and how chamber outlet ozone concentration changed over time is shown in Fig. 2. Uncertainty for ozone concentration is 1 ppb or 1% whichever is higher. Carbonyls were sampled by dinitrophenylhydrazine (DNPH) tubes and BTEX were sampled by Tenax-TA tubes. Ozone scrubbers were used 1 cm upstream of DNPH cartridges (Sigma–Aldrich, P/N 505285). Scrubbers were cleaned after each experiment following established protocols [19]. Both samples were eventually analyzed by GC–MS (Agilent 6890/MSD 5973). The GC/MS was equipped with a DB-5MS capillary column (30 m × 0.25 mm; 0.25 μm film thickness).

2.2. Building materials

Eight kinds of building materials including calcium silicate board (CSB), green calcium silicate board (GCSB), mineral fiber ceiling (MFC), green mineral fiber ceiling (GMFC), gypsum board (GB), green gypsum board (GGB), wooden flooring (WF) and green wooden flooring (GWF) were selected for this study. All selected materials are commonly used for ceiling, cabinetry, and flooring. They were unused and were shipped directly from three major manufacturers in Taiwan. Materials were wrapped in two layers of aluminum foil and one layer of plastic sheeting before experiments carried out. There is no specific manufacturer standard for conventional vs. green. But generally speaking, “Conventional” refers to building materials which are not certified by manufacturer at all while “Green” specifically refers to low emission healthy building materials (i.e., after 48 h elapsed time, the formaldehyde emission rate is less than 80 μg m^{-2} h^{-1}and BTEX emission rate is less than 190 μg m^{-2} h^{-1}) which are rated by Taiwan Architecture and Building Center (TABC) according to the requirement for green labeling evaluation program in Taiwan [31].

Each time only one kind of BM (30 cm × 30 cm) was placed and tested in the environmental chamber. The thickness is 0.9 cm, 0.6 cm, 0.9 cm, 0.9 cm, 0.7 cm, 1.1 cm, 1.2 cm and 1.2 cm for GCSB, CSB, MFC, GGB, GB, GMFC, WF, and GWF, respectively.

2.3. Carbonyls collection and analytical methods

Carbonyls were collected in dinitrophenylhydrazine (DNPH) sampling tubes at a default flow rate of 1 L/min by air pumps for 20 min (Gilian 5000). The sampling tubes were eluted with 5 mL acetonitrile, and a series of 1 μL aliquots of the eluate were then injected into GC/MS, and 13 kinds of C_1–C_8 carbonyls including formaldehyde, acetaldehyde, propionaldehyde, acrolein, butylaldehyde, crotonaldehyde, methacrolein, valeraldehyde, hexaldehyde, benzaldehyde, m-tolualdehyde, acetone and 2-butanone were analyzed by GC/MS. Although HPLC analysis is normally used in the emission testing for low molecular weight carbonyl compounds, GC/MS analysis of light carbonyls is also well established and previously used [23]. The GC/MS calibration curve for formaldehyde is provided in the Supplemental materials. The injector temperature was 300 °C; the oven temperature program was as follows: initial temperature of 50 °C for 1 min, ramp at 15 °C/min to 280 °C, hold for 1 min at 280 °C, ramp at 10 °C/min to 300 °C, hold for 5 min at 300 °C. A detector temperature of 300 °C was used for all samples. The detector was running in electron impact mode with electron energy of 70 eV and ion source temperature of 230 °C. Helium at a constant flow rate of 1 mL/min was used as carrier gas. The standard of the 13 carbonyls was purchased as a mixture of solution (CARB Method 1004 DNPH Mix2) from Supelco, USA. The mass of carbonyls was quantified by a minimum six-point external