

The Study of Carbonyl Emissions and Ozone Removal on Green Recycled Building Materials

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Abstract

Green recycled building materials (GRBMs) have the potential to reduce carbon footprint but with insufficient emission information. The increasing application of GRBMs may affect human health due to potential primary and/or secondary emissions. This study centers on assessing carbonyl emissions and ozone removal on green recycled building materials under ozone exposure circumstances. Tested materials include gypsum board (GB), waterproof gypsum board (WGB), fiber cement board (FCB), and calcium silicate board (CSB). Among all the observed carbonyls, methacrolein and formaldehyde were the only two that could be found on all four recycled building materials. All carbonyl compounds had relatively high emission factors at the beginning of the experiment and decreased to a relatively lower level one month later. Carbonyl emissions were about 33.3%, 38.9%, 42.0%, and 8.0% of their original values for WGB, FCB, CSB, and GB, respectively. The ozone removal efficiencies at the beginning and one month later were 63.26±6.11% and 61.59±5.46% on CSB, 55.74±4.94% and 47.16±7.97% on FCB, 54.59±6.50% and 34.93±6.77% on GB, and 53.63±8.40% and 23.49±6.63% on WGB, respectively. CSB seems to be a healthier recycled building material compared with WGB, GB, and FCB in terms of low carbonyl emissions and high ozone removal efficiency.

Keywords: Recycled Building Materials; Carbonyl Emissions; Ozone Removal; Formaldehyde; Methacrolein; Specific Surface Area;

1. Introduction

Indoor air quality has an enormous influence on people's health because most people spend approximately 90% of their time indoors [1]. Therefore, various air pollutants in indoor environments have been continuously investigated in a wide and diverse range, including chemical, physical, and biological contaminants [2].

Volatile organic compounds (VOCs) are among the frequently

explored indoor air pollutants. Numerous VOC species have been considered carcinogens and potentially toxic substances. For instance, formaldehyde has been proven to be a carcinogen and sensory irritant [3, 4, 5, 6]. Besides, secondary reactions between indoor VOCs and ambient ozone, especially those VOCs with unsaturated carbon bonds, from various building materials may generate or increase indoor carbonyls, such as nonanal (C9) which is irritating and can cause occupant discomfort [7, 8, 9, 10, 11].

Indoor VOCs are mainly generated internally via decoration products (i.e., paints, coatings), human activity (i.e., smoking, cooking), and furniture/building materials [12, 13]. Besides the traditional concerns for building materials based on cost, resistance to water and fire, and durability, health aspects have come to be considered in recent years. That is why wood-based panels, including particle boards, plywood panels, and medium-density fiberboard, are more commonly used as furniture and building materials [14]. Although these materials have numerous health benefits, they also have some environmental concerns. It is well known that wood itself can generate a substantial amount of formaldehyde during the manufacturing process [15]. In addition, wood composites with chemical adhesives also play an essential role as indoor formaldehyde sources [12, 16]. Thus, more environmentally friendly building materials are recommended and gradually adopted widely.

Some studies have been conducted on some green building materials regarding carbonyl emissions in laboratories. Cheng et al. [11] monitored primary emissions of carbonyls inside a test chamber and remarked that they were 75~673 $\mu\text{g}/\text{m}^2\text{hr}$ and 62~151 $\mu\text{g}/\text{m}^2\text{hr}$ at 48 hr for conventional and healthy materials, respectively. Furthermore, under the ozone concentration of 75 ppb inside the chamber, they also examined emissions of carbonyls due to secondary reactions and observed they were 7~150 $\mu\text{g}/\text{m}^2\text{hr}$ and 4~73 $\mu\text{g}/\text{m}^2\text{hr}$ for conventional and healthy materials, showing that moderate indoor ozone concentrations may cause increased concentrations of carbonyls, especially formaldehyde. Another study observed 96-hour emissions of carbonyls on four different recycled building materials in a stainless chamber, including FCB, CSB, WGB, GB, and WT (class I built wall tile) [16]. In this study, the levels of carbonyls from the highest to lowest were GB, FCB, CSB, WT, and WGB, respectively. Compared with corresponding conventional and low-VOC materials, the emissions of carbonyls from GB and CSB were 4.8 and 1.3 times those from conventional pairs and 16 and 2.5 times from low-VOC pairs.

However, studies on the carbonyl emissions from recycled materials under long-term natural ambient ozone exposure are

rare. Therefore, understanding VOC emissions, particularly carbonyl emissions associated with recycled materials' ozone reactions, is worth exploring. This work compares four commonly used recycled building materials through carbonyl emissions: gypsum board, waterproof gypsum board, fiber cement board, and calcium silicate board. For carbonyl emissions, a method of DNPH cartridges derivatization followed by GC is utilized [13, 17]. An environmental chamber was utilized to monitor the internal carbonyl emissions as a function of time on these building materials, which were exposed to the natural environment over two months when not tested. The work results reveal the health and environmental aspects of these tested materials regarding carbonyl emissions and ozone removal efficiency under real indoor environments, which helps the selection of superior recycled building materials in terms of low cost, high ozone removal, and low carbon impact.

2. Experimental Methodology

2.1 Experimental procedures

The experimental system is shown in Fig. 1. An electro-polished environmental chamber made of stainless steel was used to conduct experiments. A clean air system coupled with a flowmeter and pump provided clean air through the environmental chamber. The airflow rate, air exchange rate, temperature, and relative humidity for the environmental chamber were 1.8 L min^{-1} , 0.5 $\text{h}^{-1}\pm 3\%$, 25 $^{\circ}\text{C}\pm 0.5$ $^{\circ}\text{C}$, and 50% $\pm 3\%$, respectively. The bubble flow meter (Sensidyne, Gilibrator 2) downstream of the chamber was used to calibrate the default air flow rate. A stable amount of pure ozone was injected into the air pipeline to premix before they flew into the chamber. The inlet ozone concentration was controlled at around 60 ppb (i.e., indoor and outdoor ozone standards in Taiwan). Dinitrophenylhydrazine (DNPH) tubes were used to sample carbonyls intermittently for the first 48 hours when the building material was exposed to an ozone concentration of 60 ppb inside the chamber. After that, carbonyl samples were post-processed and analyzed by GC/FID (Agilent 7890A). A DB-5MS capillary column (30 m \times 0.25 mm; 0.25 μm film thickness) was utilized in GC/FID to quantify the amounts of carbonyls.

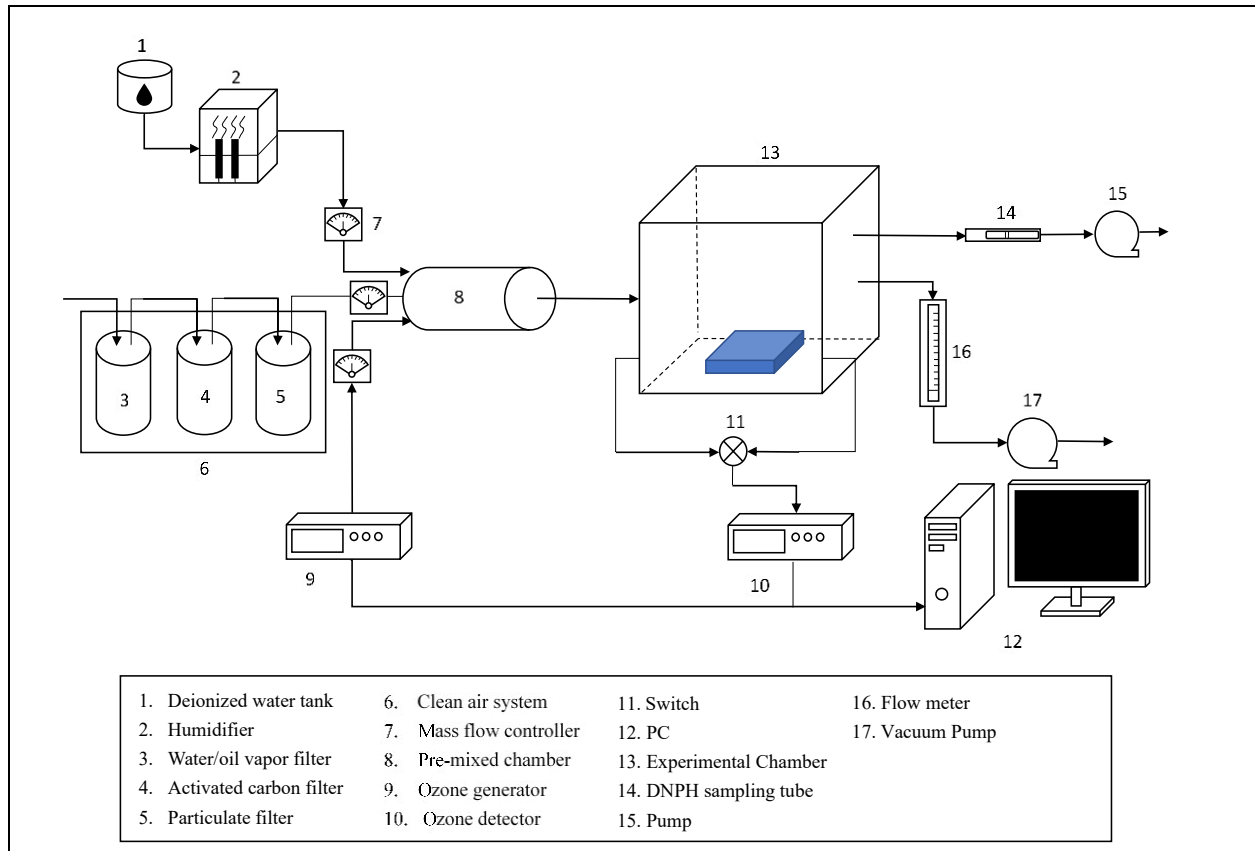


Figure 1: Experimental system

2.2 Building materials

Gypsum board (GB), calcium silicate board (CSB), fiber cement board (FCB), and water-proof gypsum board (WGB) were certified by the Taiwan Architecture and Building Center and selected for this study based on their popularity in the market.

All tested materials were purchased first-hand from manufacturers or major suppliers in Taiwan, and their contents were reported by the corresponding manufacturers in Table 1. Each material was cut into the desired size and wrapped in aluminum foil and plastic sheeting to reduce the potential emissions or reactions before running experiments.

Table 1: Contents of recycled building materials

Materials	Contents	Density (kg/m ³)
GB	Desulfurization gypsum, recycled pulp, glass fiber	600~700
WGB	Desulfurization gypsum, recycled pulp, glass fiber, waterproofing agent	600~700
CSB	Quartz powder, diatomaceous earth, cement, lime, glass fiber	800~900
FCB	Cement, Quartz sand, specially reinforced fiber, Inorganic Hybrid Materials Additives (asbestos excluded)	1200

The specimen sizes were 30 cm × 30 cm × 0.9 cm for all materials, as shown in Fig. 2. After being tested in the environmental chamber for 48 hours, each tested specimen was placed in a typical room

where windows were kept open during the daytime to simulate most residential houses in Taiwan before being rotated back to the chamber for the following experiments with one-month interval.

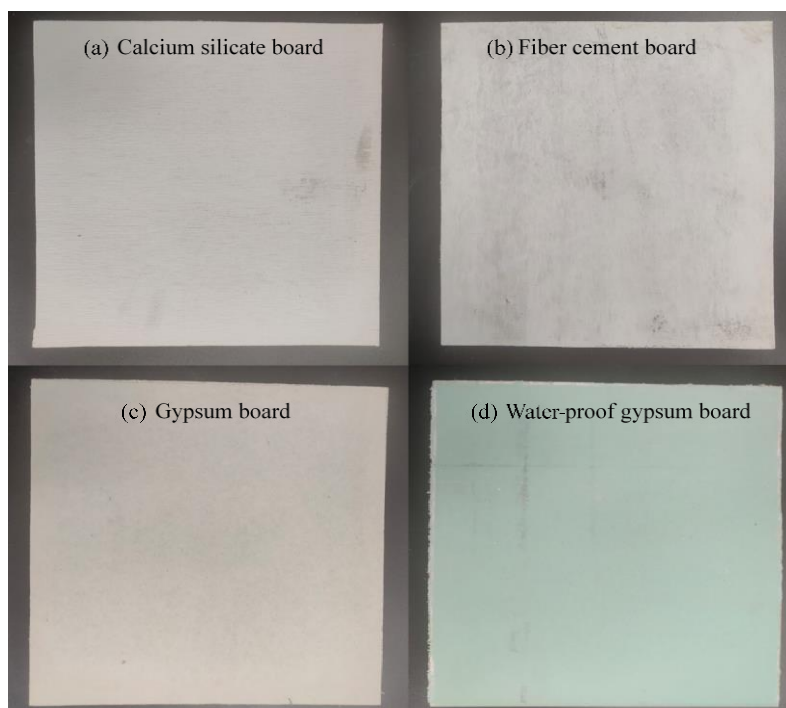


Figure 2: Specimens of the four building materials

2.3 Determination of specific surface area and total pore volume

Specific surface area and total pore volume of building material samples were obtained by Brunauere-Emmette-Teller (BET) method using BELSORP analysis software. Each sample was analyzed by automatic specific surface area and pore size distribution instrument (BELSORP-mini II). The measurement principle is based on gas volume absorbed on the solid surface of samples. The samples' specific surface area and porosity were determined by analyzing the capacity to absorb liquid nitrogen gas via a solid surface.

Each sample was degassed at 105 °C for 1 hour before analysis for BELSORP-flow II to remove the water molecules on the sample surface. Then each treated specimen of 0.1 g was measured by a microbalance sample and poured into a unique glass tube. Finally, each sample in the special glass tube was analyzed by BELSORP-mini II operation.

2.4 Carbonyls collection and analytical methods

Dinitrophenylhydrazine (DNPH) sampling tubes downstream with air pumps (Gilian 5000) were used to collect carbonyls. Each sampling event lasted 20 minutes with the default flow rate of 1L/min. After that, 5 mL acetonitrile was used to elute the sampling tubes. Then, a series of 1 µL aliquots of the eluate was analyzed by GC/FID. A six-point external calibration curve with minimum correlation coefficients of more than 0.995 was applied by GC/FID to quantify the amounts of carbonyls.

2.5 Ozone removal efficiency

The ozone removal efficiency was calculated by equation (1) as

follows:

$$\eta = \frac{(1 - C_{\text{ozone, final}})}{(1 - C_{\text{ozone, initial}})} \times 100\% \quad (1)$$

Where η is ozone removal efficiency (%), $C_{\text{ozone, initial}}$ is the initial ozone concentration, and $C_{\text{ozone, final}}$ is the final ozone concentration.

2.6 Emission Rate

Based on mass balance, the emission rates from each material were calculated by equation (2) as follows:

$$E_n = \frac{V \frac{(C_n - C_{n-1})}{(t_n - t_{n-1})} + Q C_n}{A} \quad (2)$$

Where E_n is the carbonyl emission rate normalized by area at time step n ($\mu\text{g m}^{-2} \text{h}^{-1}$), C_n is the carbonyl concentration inside the chamber at time step n ($\mu\text{g/m}^3$), C_{n-1} is the carbonyl concentration inside the chamber ($\mu\text{g/m}^3$) at time step $n-1$, Q is the air flow rate through the chamber (m^3/hr), V is the volume of the chamber (m^3), and A is the available surface area of each material for emissions (m^2).

2.7 Quality Assurance

Under the experimental conditions described above, triplicate samples of each material were tested. For the validation of the background level, the air source entering the experimental chamber was tested before each batch test. Methanol, followed by deionized water, was used to wipe and clean the inner surface of the test chamber, which was then heated until fully dry. Before the start of each experiment, the relative humidity, temperature, and

flow rate inside the chamber were stabilized after the chamber was conditioned. The MDL for individual carbonyl was 0.05 $\mu\text{g}/\text{m}^3$; any result below MDL was considered zero.

With no building materials inside the test chamber, blank carbonyl tests were performed to ensure that the background concentrations of carbonyls were under the detection limit. In addition, two control tests were performed, including the carbonyl emissions from the building materials without exposure to ozone and the ozone removal efficiency in the chamber without building materials. An infrared flow calibrator calibrated the flow rate through sampling tubes. In addition, breakthrough tests of the sampling tube were also performed to avoid the breakthrough of contaminants during sampling.

3. Results and Discussion

3.1 Ozone exposure concentration history

Results from Table 2 showed ozone exposure histories for all tested recycled building materials. The ozone concentration in the chamber was maintained at around 60 ppb during the first 48 hours, while the averaged actual indoor ozone concentration inside the room where all the test specimens were placed during the first month after the experiments for GB, WGB, CSB, and FCB were 49.84 (± 13.91) ppb, 48.90 (± 18.54) ppb, 48.50 (± 15.24) ppb, and 49.08 (± 17.20) ppb, respectively. The averaged actual indoor ozone concentration during the second month for GB, WGB, CSB, and FCB were 46.51 (± 17.36) ppb, 45.01 (± 15.28) ppb, 46.62 (± 17.59) ppb, and 45.54 (± 16.88) ppb, respectively.

Table 2: Ozone exposure concentration history for tested recycled building materials

Elapsed Time (h) Material	0~48 h (In chamber)	Real environmental ozone concentration	Around one month (Back in the chamber)	Real environmental ozone concentration	Around two months (Back in the chamber)
GB	60 ppb	49.84 \pm 13.91 ppb (48~816 h)	60 ppb (816~864 h)	46.51 \pm 17.36 ppb (864~1920 h)	60 ppb (1920~1968 h)
WGB	60 ppb	48.90 \pm 18.54 ppb (48~768 h)	60 ppb (768~776 h)	45.01 \pm 15.28 ppb (776~1486 h)	60 ppb (1486~1510 h)
CSB	60 ppb	48.50 \pm 15.24 ppb (48~768 h)	60 ppb (768~816 h)	46.62 \pm 17.59 ppb (816~1752 h)	60 ppb (1752~1776 h)
FCB	60 ppb	49.08 \pm 17.20 ppb (48~768 h)	60 ppb (768~792 h)	45.54 \pm 16.88 ppb (792~1824 h)	60 ppb (1824~1832 h)

Therefore, the daily ozone concentration in the virtual environment in this study is around 80% of the default chamber ozone concentration. It suggested that the findings here are valuable for the practical application of recycled building materials.

3.2 Carbonyl emissions from GRBMs under ozone exposure circumstances

Carbonyl emissions due to the reaction between building materials and ozone, shown in Fig. 3 to Fig. 6, were the emissions under ozone

exposure subtracted by those without ozone exposure. The results demonstrate that all carbonyl compounds have relatively high emission factors at the beginning 48 hours of the ozone exposure experiment for all tested green recycled building materials. On average, they are 40,996 $\mu\text{g}/\text{m}^2\text{hr}$ (34,531~43,374 $\mu\text{g}/\text{m}^2\text{hr}$) from waterproof gypsum board, 40,667 $\mu\text{g}/\text{m}^2\text{hr}$ (22,505~57,722 $\mu\text{g}/\text{m}^2\text{hr}$) from gypsum board, 37,483 $\mu\text{g}/\text{m}^2\text{hr}$ (31,580~43,168 $\mu\text{g}/\text{m}^2\text{hr}$) from fiber cement board, and 8,135 $\mu\text{g}/\text{m}^2\text{hr}$ (7,600~9,222 $\mu\text{g}/\text{m}^2\text{hr}$) from calcium silicate board.

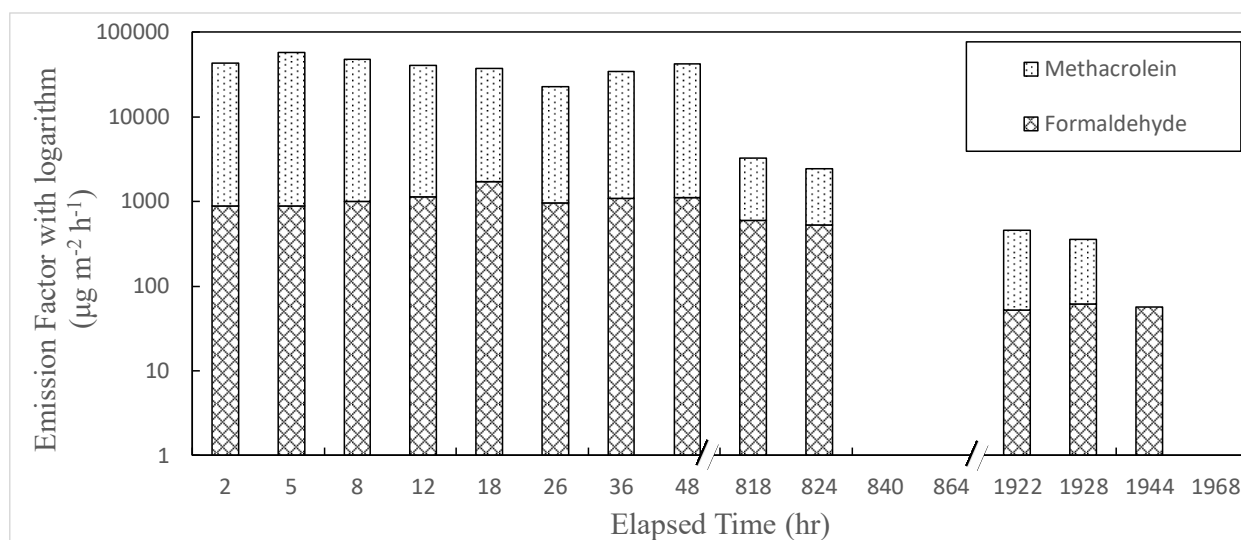


Figure 3: Emissions of Carbonyls on gypsum board versus time for the intermittent ozone exposure experiment

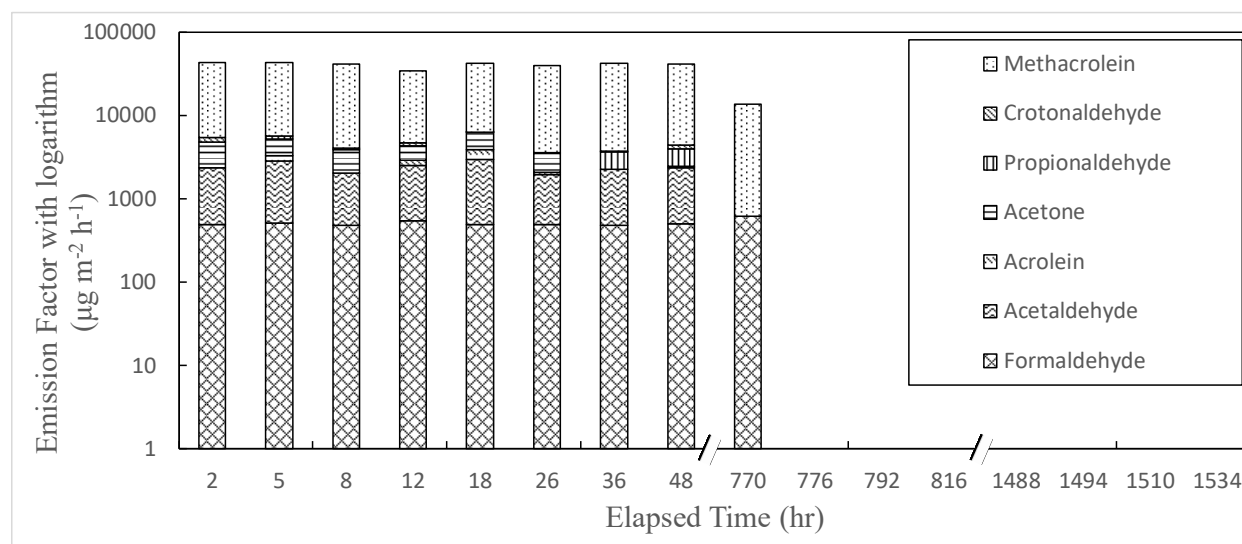


Figure 4: Emissions of Carbonyls on waterproof gypsum board versus time for the intermittent ozone exposure experiment

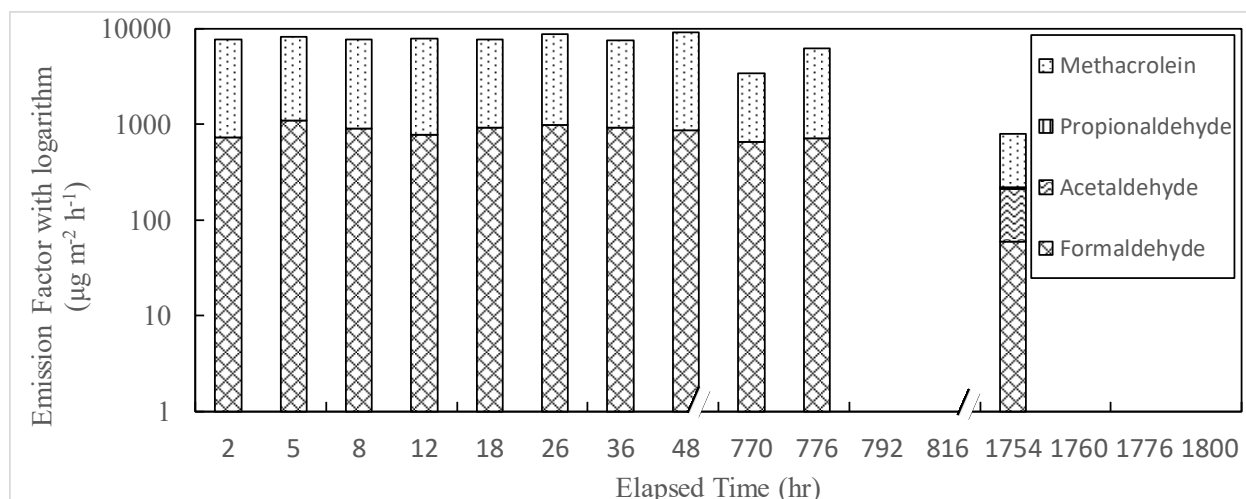


Figure 5: Emissions of Carbonyls on calcium silicate board versus time for the intermittent ozone exposure experiment

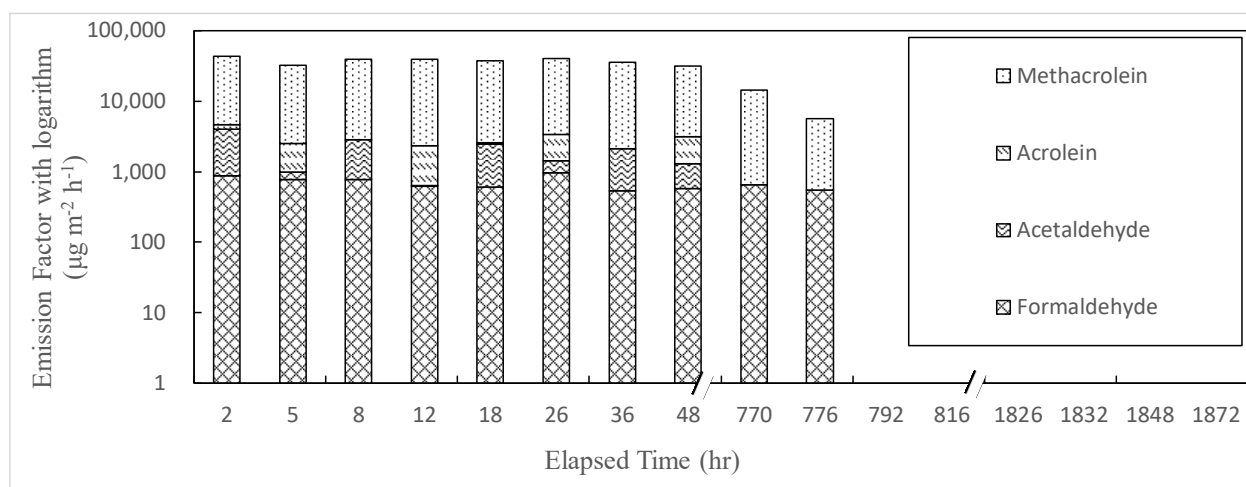


Figure 6: Emissions of Carbonyls on fiber cement board versus time for the intermittent ozone exposure experiment

One month later, their emission factors decreased to a relatively lower level. On average, they are 14,572 $\mu\text{g}/\text{m}^2\text{hr}$ from fiber cement board, 13,664 $\mu\text{g}/\text{m}^2\text{hr}$ from waterproof gypsum board, 3,418 $\mu\text{g}/\text{m}^2\text{hr}$ from calcium silicate board, 3,252 $\text{g}/\text{m}^2\text{hr}$ from gypsum board. They are about 38.9%, 33.3%, 42.0%, and 8.0% of the original values, respectively. Two months later, they dropped much more; some even dropped to zero for FCB. They are only 9.8%, 1.3%, and 1.1% of the first-month data for CSB, WGB, and GB, respectively. These emissions detected after two months were mostly found at the first sampling, so it was suspected that some organic compounds from dust or human activity accumulated during the second month's ambient exposure [18]. These limited amounts of organic compounds on the surface of each material reacted with ozone so quickly that the carbonyl emissions were observed only for the first sampling but then became too low to be detected.

It has been observed that there were seven types of carbonyl compounds detected from recycled green building materials

when materials were exposed to an ozone environment; observed carbonyls including formaldehyde, acetaldehyde, acrolein, acetone, propionaldehyde, methacrolein, and crotonaldehyde. The species of carbonyl emissions were the largest from the waterproof gypsum board, covering all seven carbonyls compounds; the second largest from calcium silicate board and fiber cement board, covering four carbonyls compounds; the least from the gypsum board, only covering two carbonyls.

Among all the observed carbonyls, methacrolein and formaldehyde are the only two that could be found on all four recycled building materials. Many research studies determined that ozone reactions with isoprene which widely exists in all kinds of wood products, mainly formed methacrolein and formaldehyde with yields ranging from 0.34–0.42 and 0.85 to 0.96, respectively [19, 20, 7]. In this study, the relative percentage of methacrolein was consistently the largest among all the carbonyls detected in each experiment, ranging from 72.42% to 100%. Compared with those previous studies above, a higher level of methacrolein than formaldehyde

here may be caused by the differences in ozone concentration, 60 ppb in this study vs. 1000 ppb in previous studies. In other words, given that methacrolein and formaldehyde are both byproducts of ozone reactions, ozone may further react with methacrolein at higher ozone concentrations, which leads to less methacrolein and more formaldehyde. As for formaldehyde, the emission factors at the beginning and one month later were 881-1,131 $\mu\text{g}/\text{m}^2\text{hr}$ and 525-596 $\mu\text{g}/\text{m}^2\text{hr}$ from GB, 481-549 $\mu\text{g}/\text{m}^2\text{hr}$ and 0-717 $\mu\text{g}/\text{m}^2\text{hr}$ from WGB, 780-1,109 $\mu\text{g}/\text{m}^2\text{hr}$ and 662-718 $\mu\text{g}/\text{m}^2\text{hr}$ from CSB, 542-975 $\mu\text{g}/\text{m}^2\text{hr}$ and 558-648 $\mu\text{g}/\text{m}^2\text{hr}$ from FCB. It seems that the emissions of formaldehyde decrease as time goes by. This is likely because formaldehyde commonly existed in all test materials, and they were detected mainly as primary emissions rather than secondary reaction products. Thus, over time, there is less and less formaldehyde inside these materials. However, the relative percentage of formaldehyde among all carbonyls detected one month later increased when compared with the initial period of the experiment. The relative percentages of formaldehyde among all carbonyls at the beginning and one month later are 2.70% and 19.76% for GB, 1.22% and 4.49% for WGB, 11.16% and 14.33% for CSB, 1.92% and 5.94% for FCB (Fig. 2-5), respectively. The reason is suspected that formaldehyde from primary emissions decreased with time and eventually became less important than that from secondary reactions.

Methacrolein is moderately toxic and is mainly used in copolymer and resin manufacturing. Formaldehyde is a well-known carcinogen. The results show that methacrolein and formaldehyde were consistently detected throughout the experimental period. Thus, it is highly suggested that some preventative actions be taken to protect occupants or workers, at least during the first month when these materials, especially WGB and GB, are used.

3.3 Ozone Removal on GRBMs

From Table 3, the ozone removal efficiencies at the beginning and one month later are 63.26±6.11% and 61.59±5.46% on CSB,

55.74±4.94%, and 47.16±7.97% on FCB, 54.59±6.50% and 34.93±6.77% on GB, and 53.63±8.40% and 23.49±6.63% on WGB, respectively. The ozone removal efficiency observed two months later became 50.59±4.23% on CSB, 47.27±7.45% on GB, 19.23±6.32% on WGB, and 7.26±3.73% on FCB, respectively. It can be seen that ozone removal efficiencies on all tested materials generally decreased with time elapsed. However, the two-month ozone removal efficiency of 47.27% seemed to be recovered compared with the one-month data of 34.93% on GB. The reason is suspected to be the diffusion of ozone-reactive compounds inside out to the surface, which still needs further exploration. Among all the test materials, CSB showed consistently high ozone removal efficiency, which remained at 80% of the original after two months. This is supposedly related to the high specific surface area. Lin and Hsu [21] observed that CSB, with the highest surface area and total pore volume, showed the highest ozone removal efficiency among eight conventional and green building materials. They concluded that specific surface area has a strong positive correlation with ozone removal, which is highly comparable to the results of this study. Table 4 compares this study's specific surface area and ozone removal against the previous study on similar building materials. It is observed that ozone removal efficiencies were 62% and 64% on conventional CSB and green CSB, quite similar to the initial 63% on recycled CSB in this study. However, their ozone removal efficiencies were 40% and 50% on conventional GB and green GB, lower than the initial 55% on recycled GB in this study. This can be derived from the similar trend of specific surface area, which is 27.25 m^2/g and 34.87 m^2/g on conventional CSB and green CSB in the previous study vs. 27.62 m^2/g on recycled CSB in this study. As for GB materials, they are 1.11 m^2/g and 1.29 m^2/g on conventional GB and green GB in the previous study, lower than 1.84 m^2/g on recycled GB in this study. The relationship between the specific surface area and ozone removal is derived and shown to be positively strong (Fig 7) with $R^2=0.9756$.

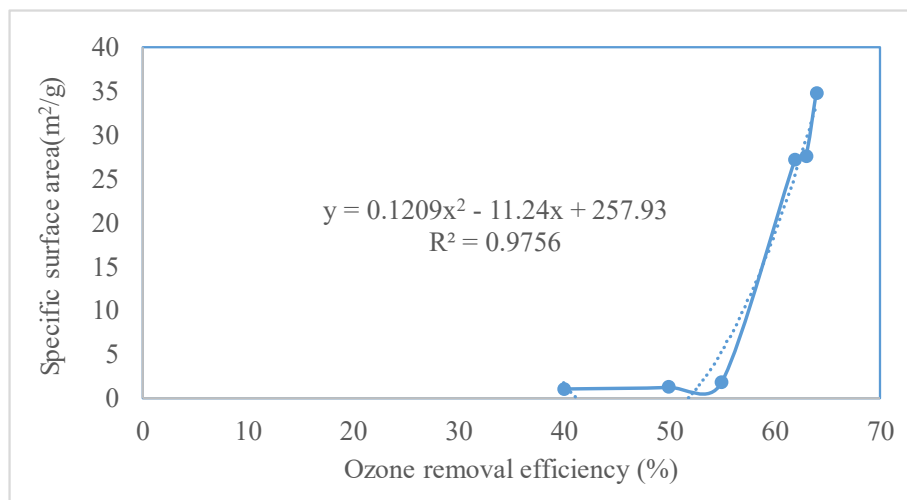
Table 3: Ozone removal efficiency on green recycled building materials

Material \ Elapsed Time (h)	The first 48 hours	Around one month (Back in the chamber)	Around two months (Back in the chamber)
GB	54.59±6.50%	34.93±6.77% (818~840 h)	47.27±7.45% (1922~1968 h)
WGB	53.63±8.40%	23.49±6.63% (770~776 h)	19.23±6.32% (1488~1510 h)
CSB	63.26±6.11%	61.59±5.46% (770~792 h)	50.59±4.23% (1754~1776 h)
FCB	55.74±4.94%	47.16±7.97% (770~792 h)	7.26±3.73% (1826~1832 h)

Table 4: Comparison of the specific area and ozone removal on GB and CSB

Building material	Specific area (m ² /g)	O ₃ removal efficiency (%)
Recycled GB	1.84	55
Conventional GB*	1.11	40
Green GB*	1.29	50
Recycled CSB	27.62	63
Conventional CSB*	27.25	62
Green CSB*	34.87	64

Note: *refers to the results of the previous study by Lin and Hsu (2015)

**Figure 7: Relationship between ozone removal and specific surface area**

The ozone removal efficiencies estimated according to the specific area of WGB and FCB are 53.01% and 61.4%, while the measured ozone removal efficiencies are 53.63% and 55.74%. Thus, WGB shows the same trend as CSB and GB again. However, FCB shows a lower ozone removal than predicted. The reason is not entirely certain, but it is highly suspected because a larger portion of FCB contents is inorganic, including cement, and inorganic hybrid materials.

4. Conclusions

1. The results above showed that CSB could be a potentially healthier recycled building material compared with WGB, GB, and FCB in terms of low carbonyl emissions and high ozone removal efficiency.
2. Methacrolein has the most significant emission among all the detected carbonyls throughout the ozone exposure experiments, probably due to secondary reactions between ozone and unsaturated organic compounds in building materials. More stringent testing guidelines for recycled building materials about potential secondary emissions related to ozone exposure are highly recommended.
3. WGB and GB showed the highest carbonyl emissions under ozone exposure, indicating that more stringent tests will be conducted on recycled GB before they are sold and applied indoors.

4. Formaldehyde emissions were detected for all tested recycled building materials under a typical actual environment. Thus, preventative strategies such as retaining a sufficient amount of continuous ventilation, especially during the first month, are highly recommended.

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Author Contributions

Yu-Hsiang Cheng: Data curation. Chi-Chi Lin: Funding acquisition, Supervision, Conceptualization, Writing - review & editing. Kai-Jyun Ke: Material preparation and Data collection.

Ethical Approval

Not applicable.

Consent to Participate

Not applicable.

Consent to Publish

The manuscript is reviewed and approved by all authors.

Availability of data and material

All relevant data and materials are visible in the manuscript.

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