Formaldehyde released in leachate from medium density fiberboard (MDF) buried in a simulated landfill

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Formaldehyde, a flammable, colorless, highly reactive gas at standard temperature and pressure, is commonly found in the environment, however, it is toxic and causes health issues for humans. Approximately 14 million tons of wood waste containing formaldehyde based resins are generated yearly and disposed in landfills or burned. No regulations exist and no studies have been conducted to address formaldehyde emission from wood waste containing formaldehyde buried in landfills. Studies are therefore needed to address this potential environmental issue. The objective of this study was to determine the amount of formaldehyde released into the leachate from two sizes of medium density fiber board (MDF) buried in a simulated landfill. Simulated landfills were constructed in cylindrical plastic containers (15.24 cm diameter, 22.86 cm high) with alternating layers of silty clay soil and ground or cut pieces of MDF for a total of five layers. Leachate was collected and sampled for formaldehyde and pH on days 0, 7, 14, 21 and 28. Formaldehyde released in leachate was determined by derivatizing using 2,4-Dinitrophenylhydrazine and analyzing by liquid chromatography with UV-Vis detection. Preliminary results indicate that formaldehyde released in the leachate was reduced by 99% by the end of the study. The initial pH of the leachate from soil without MDF was 5.87 and increased to 6.18 at the first week's sampling time and remained at approximately 6.22 through day 28. The leachate from soil with added MDF had an initial pH of 4.66 and increased weekly to 6.40 on week four. Results from this study should provide new information about the fate of wood waste containing formaldehyde disposed in landfills.

Introduction

The forest products industry was developed to meet the building needs of humans and has grown worldwide (Youngquist et al. 1996). Many forests have been destroyed by reckless clear-cutting practices causing people to be concerned about the future of forest, wildlife diversity, wood production, and the aesthetics (McNutt et al. 1992). However, current supplies of forest products materials are not sufficient to meet ordinary building needs. Therefore, wood composites were developed mostly during the past 40 years to meet the needs of the forest products industry (Thomas et al. 2008). In 1996, millions of tons of wood composites were manufactured annually (Maloney 1996). In 2009, the United Nations estimated that 42,494 m^3 of wood-based panels were used (Pepke 2010).

Wood composites are defined as wood that has been bonded and compressed with an adhesive (Maloney 1996) and include products such as: medium density fiberboard (MDF), particleboard (PB), oriented strand board (OSB), plywood, and laminated beam. Many adhesives commonly used in wood composite production contain formaldehyde: melamine-formaldehyde (MF), phenol-formaldehyde (PF), melamine-urea-formaldehyde (MUF), and urea-formaldehyde (UF). Formaldehyde is not only widely used in the manufacture of wood products such as medium density fiberboard (MDF), oriented strand board

(OSB), plywood, laminated beam, and furniture but also in the chemical and paper industries as well as in textile processing (Marra 1992). All resins are thermosetting and once formed they become very stable and resistant to heat. Among the different types of adhesives, UF resins are widely used in industry because of their beneficial characteristics. UF resins cost less than other resins and penetrate wood cells very quickly. Urea-formaldehyde resins can be degraded by bacteria into urea, formaldehyde, ammonia and carbon dioxide (Jahns et al. 1998)

Formaldehyde, a flammable, colorless, highly reactive gas at standard temperature and pressure, is commonly found in the environment (IPCS 2002). In nature the formaldehyde concentration is less than 1 μg/m^3 with an average value of 0.5 μg/m^3 (IARC 1995). However, formaldehyde is toxic and causes health issues such as watery eyes, burning sensations in the eyes and throat, nausea, difficulty in breathing and cancer in humans (IARC 2006).

In 1996, the World Health Organization established the drinking water quality guideline value of 0.9 mg/liter for formaldehyde and air quality guideline value of 0.1 mg/m^3 (IPCS 1996). The U.S. Department of Labor's Occupational Safety and Health Administration (OSHA) established a standard of 0.75 parts per million for 8 hours working time with appropriate labels and warnings to protect workers from exposures to formaldehyde (OSHA 2011). The U.S. Environmental Protection Agency (EPA), Consumer Product Safety Commission (CPSC), and the U.S. Department of Housing and Urban Development (HUD) have focused on indoor air formaldehyde exposure. On July 7, 2010, the Formaldehyde Standards for Composite Wood Products Act was signed into law which states that no higher than 0.05 parts per million of formaldehyde for hardwood plywood, or 0.06 parts per million of formaldehyde for particleboard and MDF are allowed for emission of formaldehyde and this law will become effective on January 1, 2013 (S. 1660-6).

Approximately 14 million tons of wood waste containing formaldehyde based resins are generated yearly and disposed in landfills or burned (EPA 2003). Formaldehyde bonded wood waste may be a source of formaldehyde emission by release of free formaldehyde through degradation of the urea-formaldehyde bond in wood waste. No regulations exist and to our knowledge no studies have been conducted to address formaldehyde emission from formaldehyde bonded wood waste buried in landfills. Therefore the objective of this study was to determine the amount of formaldehyde released in leachate by two sizes of MDF buried in a simulated landfill.

Materials and Methods

Experimental design

Simulated landfills were constructed in cylindrical plastic containers (15.24 cm diameter, 22.86 cm high) with alternating layers of silty clay soil (870 g) and MDF (120 g, ground or cut pieces) for a total of five layers (Figure 1). Soil (silty clay) was collected in Starkville, Mississippi and was obtained from a depth of 152.4 cm and then sieved through a screen to remove debris and large rocks. Plastic screens (10 mm thick and 5 mm thick), and non-woven fabric were placed successively on the bottom of each container in order to prevent clogging of the collection tube. Non-woven fabric was also placed on the top soil layer in order to reduce loss of moisture. One circular hole (5 mm diameter) was drilled in the bottom of the containers for collection of leachate and two circular holes (5 mm diameter) were drilled in the top of each plastic cylindrical container for air sampling. Plastic tubing containing a cut-off valve was attached through each hole with glue for air sampling and leachate collection. All chambers were stored in an incubator at 34°C.

Medium density fiberboard

Medium density fiberboard (MDF, 100 cm x 100 cm x 1.27 cm), commercially manufactured was used in this study. Two different MDF sizes were tested: cut pieces (3 cm x 1.5 cm x 0.5 cm) and ground (milled though a 5 mm screen). There were four treatments with three replicates per treatment: 1) ground MDF covered in soil, 2) cut pieces of MDF covered in soil, 3) soil only, and 4) ground MDF only.

Leachate collection and sampling

Deionized water (750 mL) was added to each constructed landfill initially to saturate the soil and MDF, then drained (by gravity) into a glass vial, filtered though a 0.22 µm Nylon filter and analyzed for formaldehyde (day 0 sample). A second portion of deionized water (200 mL) was added to the chambers and allowed to soak for 1 hour. After 1 hour, leachate was collected in a 50 mL glass vial and filtered through a 0.22 µm Nylon filter (day 1 sample). At day 7 and on subsequent sampling times, 200 ml of deionized water was added to each treatment and allowed to soak for 1 hour and then collected as described above. Leachate was sampled on days 0, 7, 14, 21 and 28.

Formaldehyde determination

Formaldehyde was analyzed according to the U. S. Environmental Protection Agency Method IP-6C and 8315A (US EPA 1996a, US EPA 1996b) using a Waters 2695 high-performance liquid chromatography system with UV-Vis detection at 370 nm (Waters 996, Waters Corporation, Milford, MA). The analytical column was a 3.9 x 150 mm HPLC column (Nova-Pac® C18 60Å 4 µm, Waters Corporation, Milford, MA). HPLC chromatographic conditions were as follows: 40/60 acetonitrile/water (v/v), hold for 1 min; 40/60 acetonitrile/water to 100% acetonitrile in 3 min; 100% acetonitrile for 10 min; Flow Rate: 1.0 mL/min; Injection Volume: 20 µL.

Leachate (1 ml) was derivatized by adding 0.5 mL of 2, 4-dinitrophenylhydrazine (DNPH) solution (100 mg/ 100mL in 0.1 N HCl) and mixing for 20 minutes. Derivatized formaldehyde was extracted with 1 mL of toluene, evaporated to dryness under nitrogen, resuspended in 1 mL acetonitrile and injected into the HPLC-UV system. The volume of leachate collected and pH were also determined at each sampling time.

Formaldehyde concentration was determined from a calibration curve generated using 0, 0.25, 1, 10, 25 and 100 ppm derivatized formaldehyde standards in acetonitrile. The formaldehyde concentration in the leachate was converted to total micrograms formaldehyde by multiplying the concentration determined from the calibration curve by the volume of leachate collected.

Results

Formaldehyde analysis

The amount of formaldehyde (µg) in the leachate from the four treatments were determined weekly for 4 weeks. The highest amount of formaldehyde was observed on day 1 and occurred in all treatments after the collection of the saturation leachate (day 0, Figure 2). On day 1 the leachate from MDF only (treatment 4) was highest in formaldehyde (43 mg) compared to MDF plus soil (17 and 15 mg respectively, treatments 1 and 2) while the soil only treatment (# 3) contained the lowest amount of formaldehyde (0.5 mg) in the leachate. The amount of formaldehyde decreased by 50% in the ground MDF only treatment by day 7 compared to between 90-96% in the MDF plus soil treatments (1 and 2) and the soil only treatment. The amount of formaldehyde in each treatment continued to decrease on days 14 and 21. By the end of the study at day 28, the formaldehyde in the leachate had been reduced by 99% in treatments 1 and 2 and by 87% in the MDF only leachate (treatment 4).

pH and leachate volume

The change in pH of the leachate from the 4 treatments over the four week test period is shown in Figure 3. The initial pH of the leachate from soil without MDF was 6.36, increased to a maximum of 6.87 at day 21 and was 6.70 at day 28. The leachate from ground MDF buried in soil (treatment 1) had a pH of 4.66 on day 0 and increased to 6.70 by day 28. At day 28 the pH of the leachate containing the small MDF was 6.56 which was slightly lower than the pH of the leachate from the ground MDF in soil. The pH of the ground MDF only treatment was 4.21 on day 0 and increased to 5.71 on day 28 which was lower than the leachate in the other treatments.

The highest change in leachate collection volume was a decrease in the MDF only treatment that occurred at the day 1 sampling. After day 1 the volume of collected leachate remained stable in this treatment. In contrast, the volume of leachate collected in the treatments containing soil and MDF increased at day 1, decreased at days 7, 14 and 21 then increased at day 28. The volume of leachate collected in the soil only treatments decreased from day 0 to day 1 and remained constant after day 1. Figure 4 shows the leachate amounts of the four treatments at each sampling time.

Discussion

The pH of the leachate was lower on ground and small pieces MDF buried in soil and the ground MDF only treatments (1, 2, and 4 respectively) than in the soil only treatment (3) except at the end of the study. This is most likely due to the acidic catalysts used in making the UF resin and the acidic wood fibers. However, the pH of the leachate increased in treatments 1 and 2 to that of the soil's pH by the end of study. The pH of the ground MDF only (treatment 4) remained below that of the other treatments, indicating that the soil helped to neutralize the acidity of the leachate overtime. This data indicates that most of the acidic material in the MDF plus soil treatments was removed during the 4 week test period.

The amount of leachate collected from soil only (treatment 3) was more constant over time compared to the other treatments containing MDF. At the last sampling time, the amount of leachate from the treatments containing MDF plus soil (1 and 2) increased 4-5x.

The formaldehyde in leachate (Figure 2) from ground MDF only (treatment 4) was reduced by 50% by the second sampling time and this reduction increased to 83% by the end of the study. The formaldehyde reduction in MDF plus soil treatments increased to 90-96% at day 7 and increased again to 99% at the end of the study. This indicates that formaldehyde may be bound to the soil thereby reducing its concentration in the leachate over time.

At the end of the study, the amounts of formaldehyde in the leachate from ground MDF buried in soil and small pieces MDF buried in soil were less than the formaldehyde in leachate from soil only on Day 0. From this data, nearly all of the free formaldehyde was removed from the MDF plus soil treatments. However, formaldehyde was still detected in the leachate from ground MDF only. Reduction of formaldehyde in the leachate from soil amended treatments may have been due to transformation or degradation of formaldehyde by the soil or binding to the soil. Further studies are needed to determine the amount of formaldehyde in the soil and remaining in the MDF and to address the fate of the formaldehyde in soil.

Acknowledgements

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Figure 1. Simulated landfill test design for determination of formaldehyde in leachate and air. A: Leachate port (5mm), B: Plastic screen (10mm), C: Plastic screen (5mm), D: Non-woven fabric, E: Soil (2.54 cm), F: MDF sample (2.54 cm).

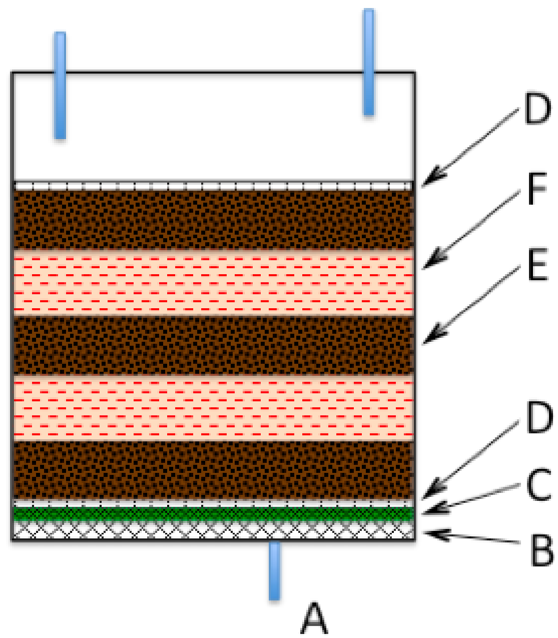

Figure 2. Total formaldehyde (µg) released in leachate from 4 treatments involving MDF buried in a simulated landfill and sample weekly for 4 weeks. The concentration represents the average values of three replications.

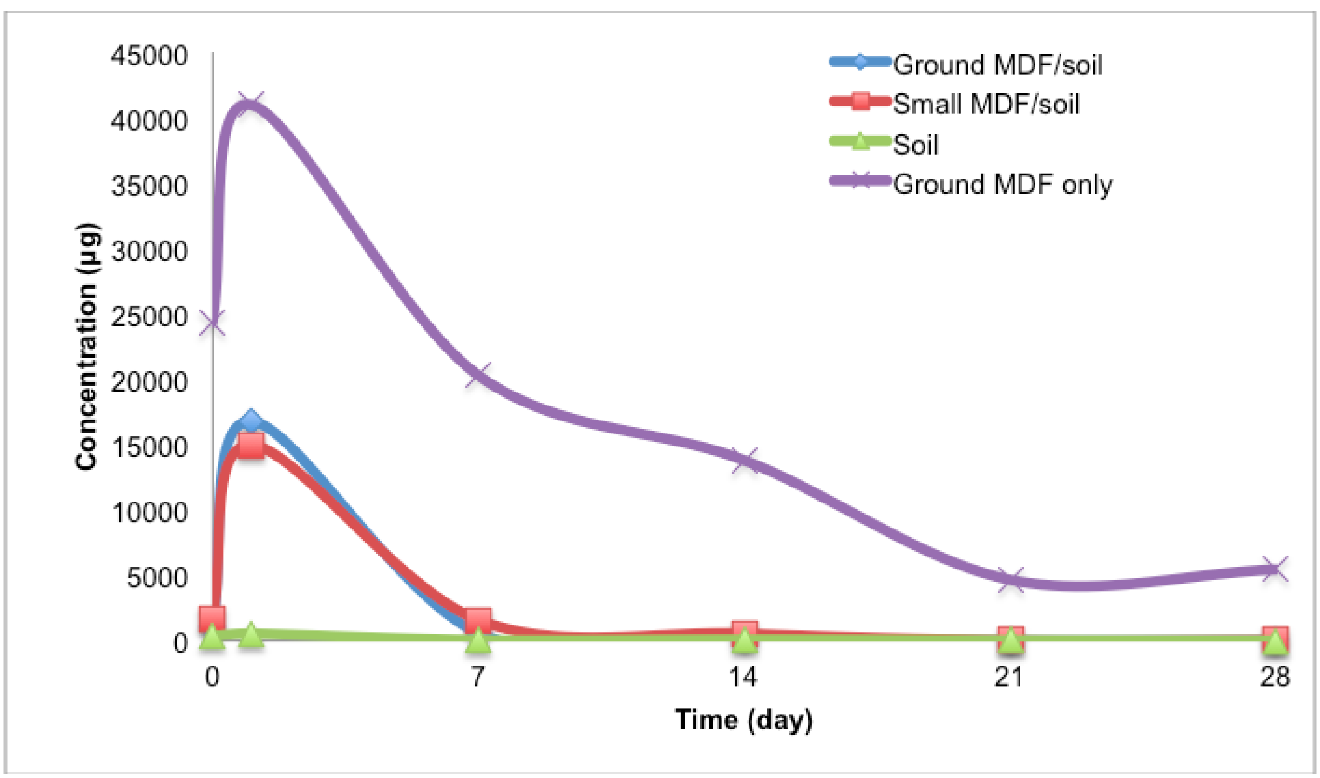

Figure 3. pH of leachate from 4 treatments involving MDF buried in a simulated landfill and sampled weekly for 4 weeks. The pH represents the average of three replications.

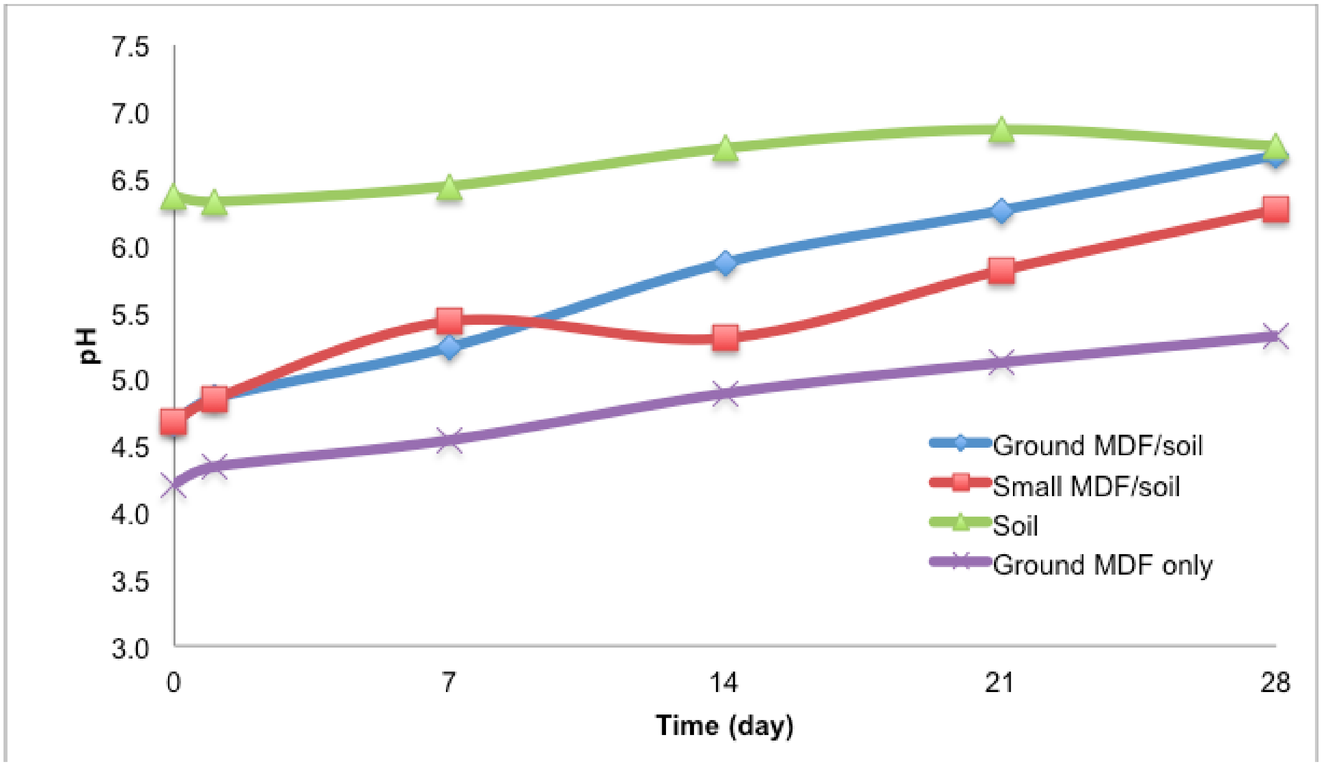

Figure 4. Volume of leachate collected from 4 treatments involving MDF buried in a simulated landfill and sampled weekly for 4 weeks. The volume represents the average of three replications.