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Formaldehyde Release from Medium Density Fiberboard in Simulated Landfills for Recycling[1]

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ABSTRACT

Laboratory-scale landfills (simulated landfills) were designed to determine the formaldehyde released into air and leachate from medium density fiberboard (MDF). Simulated landfills were constructed using cylindrical plastic containers containing alternating layers of soil and MDF for a total of five layers. The highest concentration of formaldehyde was found in the air and leachate from the MDF only treatment compared to treatments containing MDF and soil. At the end of the study (28 days), formaldehyde concentrations in air and leachate from treatments containing MDF and soil decreased by 70 percent and 99 percent, respectively, while the treatment containing MDF only still released formaldehyde into the air and leachate. Therefore, waste MDF after storing 4 weeks in water may be recycled as compost or mulch based on formaldehyde leaching. Also, these data indicate soil restricts formaldehyde release into air and leachate and provides new information about the fate of wood-based composite waste containing UF resin disposed in landfills.

Keywords : simulated landfills, medium density fiberboard, formaldehyde, leachate, urea-formaldehyde resin, landfill

1. INTRODUCTION

Reclamation, which means recycling and resource recovery, has been a biggest assignment for mankind due to the exhaustion of natural resources. Wood products are one of the candidates of recycling. The wood products industry was developed to meet the building needs of humans and has grown worldwide (Youngquist *et al*. 1996). Wood composites were developed mostly during the past 40 years to meet the needs of the forest products industry and now are a viable option (Thomas *et al*. 2008). In 1996, several million tons of wood composites were manufactured annually in USA (Maloney 1996). Wood composites are defined as wood that has been bonded and compressed with an adhesive and include products such as: medium density fiberboard (MDF), particleboard (PB), oriented strand board (OSB), plywood, and laminated beam. Among the different types of adhesives such as melamine-formaldehyde (MF), phenol-formaldehyde (PF), melamine-urea-formaldehyde (MUF), and urea-formaldehyde (UF), UF resins are

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widely used in MDF and PB manufacture because of their beneficial characteristics of lower cost and easier handling.

Composite wood products are susceptible to decay by microorganisms (Chung *et al*. 1999). UF resins can be degraded by bacteria into urea, formaldehyde, ammonia, and carbon dioxide (Jahns *et al*. 1998). Therefore, most wood waste containing formaldehyde-based resins is disposed in landfills or burned (US EPA 2005). On the other hands, famers used sawdust as mulch to improve soil condition and prevent erosion (EPA 2011). However, MDF sawdust is currently not being used in agriculture due to the lack of understanding of the environmental fate of formaldehyde resins from MDF, even though a previous study found that formaldehyde was not detected in mulch made from MDF sawdust (EPA 2011).

Unfortunately, disposing of wood-based composite wastes in landfill requires to consume valuable land space and economical cost, and to prevent contamination from wood waste. Specifically, the rule of California does not allow, therefore, disposal of wood composites containing formaldehyde-based resins because of environmental concerns (No. 27 CA code of regulation). Wood waste containing UF resins may be a source of formaldehyde emission by the release of free formaldehyde or degradation of the UF resin in wood waste although formaldehyde can be efficiently reduced by anaerobic microorganisms (Lu and Hegemann 1998). No regulations exist for formaldehyde emission from wood waste containing UF resin buried in landfills and yet the formaldehyde released into air or leachate after disposal of the wood wastes in landfills is not known.

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 ug/m^3 with an average value of 0.5 ug/m^3 (IARC 1995). At certain levels, however, formaldehyde is toxic and causes health issues such as eye or skin irritation (0.1 - 5 ppm), difficulty in breathing (5 - 20 ppm), and lung irritation or death in severe cases (20 - 100 ppm). Long time exposure can also cause cancer (IARC 2006). Therefore, formaldehyde emission has been regulated by many government organizations or wood products association. For example, the World Health Organization established the drinking water quality guideline of 0.9 ppm for formaldehyde and air quality guideline of 0.1 mg/m^3 (IPCS 1996). The new formaldehyde guidelines are 0.05 ppm for plywood, and 0.09 ppm for PB and MDF in the USA (S.1660-6 2010). However, all current regulations are for in-door applications or workplace exposure.

In the pilot study (EPA 2011), the UF resin decomposed to harmless end products such as carbon dioxide, ammonium and water, and the amount of formaldehyde released in the air from ground-up MDF waste was still unknown (EPA 2011). Unfortunately, determination of formaldehyde in air and leachate directly from a landfill is not simple because of restrictions on landfills including sufficient amounts of leachate or air collections and environmental factors.

In order to recycling of medium density fiberboard (MDF), there are two approaches: re-manufacturing to fiberboard with waste wood products and using as mulch. However, two approaches considered low cost efficiency and the lack of understanding of environmental issue. Most wood waste containing urea-formaldehyde (UF) resin, therefore, is disposed in landfills or burned. No studies have been conducted to determine formaldehyde released from wood waste buried in landfills. The objective of this study was to determine the formaldehyde in air and leachate released from MDF buried in simulated landfills as a possible way of recycling.

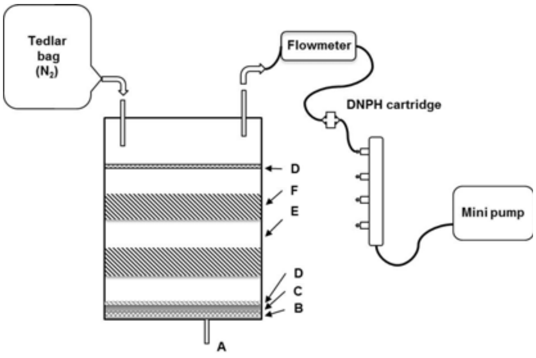

Fig. 1. Simulated landfill design for determination of formaldehyde in leachate and air. A: Leachate port (5 mm), B: Plastic screen (10 mm), C: Plastic screen (5 mm), D: Non-woven fabric, E: Soil (2.54 cm), F: MDF sample (2.54 cm).

2. MATERIAL and METHODS

2.1. Materials

MDF (100 cm × 100 cm × 1.27 cm) used in this study was commercially manufactured (Georgia-Pacific, NC) and purchased approximately 10 years ago and stored at Forest Products Department at Mississippi State University. Two sizes of MDF were prepared: cut pieces (3 cm × 1.5 cm × 0.5 cm) and ground (milled though a 5 mm screen). Soil (silty clay) was collected in Starkville, Mississippi and then sieved through a screen (0.5 cm) to remove large rocks. A 2,4-dinitrophenylhydrazine (DNPH) cartridge was purchased from Waters Corporation (Milford, MA). DNPH, formaldehyde solution (37 percent in organic-free reagent water), hexane, and acetonitrile (HPLC grade) were purchased from Sigma-Aldrich Co. (St. Louis, MO).

2.2. Simulated Landfill Design

Simulated landfills were constructed in plastic containers (15.24 cm diameter, 22.86 cm high) and alternating layers of soil (total 870 g) and MDF (total 120 g, ground or cut pieces) for a total of five layers (Fig. 1). Each layer was distinctively separated but contacted each other. Two sizes of plastic screen (10 and 5 mm thick, respectively) and non-woven fabric were placed successively on the bottom of each container and non-woven fabric was placed on top layer of soil. One hole (5 mm diameter) was drilled on the bottom side of the containers for leachate collection and two holes (5 mm diameter) were drilled on the top of each plastic lid for air sampling. Plastic tubing containing a cut off valve was attached through each hole with glue for air sampling and leachate collection. All containers were stored without lid in an incubator at 34℃.

2.3. Air Sampling

At the sampling time, containers were closed with lids and then air in the headspace of each container was collected before adding water on days 0, 7, 14, 21 and 28 as described in Fig. 1. A Tedlar bag (25 L, TP-25, TRS environment™, Chesterfield, MI) filled with nitrogen (ultrapure grade) was connected to the inlet tube in the top of the container. The outlet tube was connected to a flowmeter and then to a DNPH cartridge. The cartridges were connected to a manifold which was connected to a mini-pump (Gilian 3500, Sensidyne®, Clearwater, FL) set to pull headspace air from the container for 4 hours at 60 mℓ per minute. Formaldehyde was trapped and derivatized in the DNPH cartridge. Collected amount of formaldehyde was generated from each sample at the sampling time (4 hours and total 24 L) rather than accumulated amount of that.

2.4. Leachate Sampling

After air sampling, deionized water (750 mℓ) was

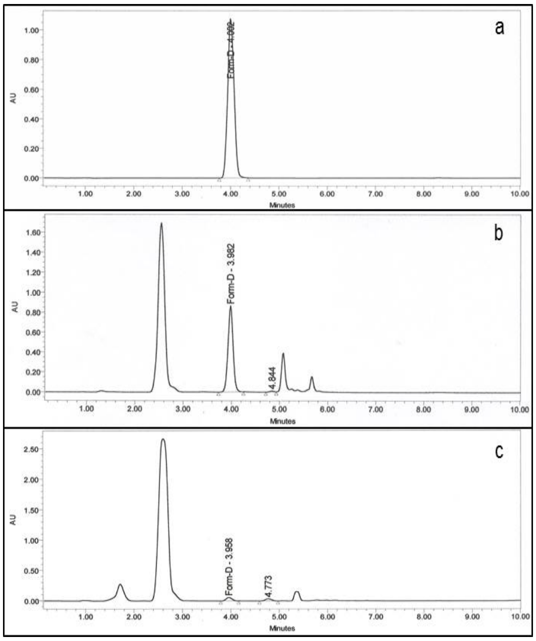

Fig. 2. HPLC analysis of formaldehyde-DNPH standard (a), leachate (b), and air (c) at 370 nm.

added first to saturate the soil and MDF, and then drained and collected on day 0 only. A second portion of deionized water (200 mℓ) was added to the container and allowed to soak for 1 hour. After 1 hour, leachate was collected and filtered through a 0.22 μm nylon filter. Leachates were sampled weekly.

2.5. Synthesis of Formaldehyde-DNPH Standard

DNPH (0.4732 g) was dissolved in 2N HCl (150 mℓ) to obtain an approximately saturated solution. Formaldehyde (0.3 mℓ) was added to the DNPH solution. The resulting precipitate of formaldehyde-DNPH was collected and filtered through a 0.45 μm polypropylene membrane. The derivatized formaldehyde with DNPH was recrystallized in acetonitrile and then vacuum dried. In this experiment, the melting point obtained for formaldehyde-DNPH was 154 - 155°C, and one clear yellow spot was visible on the thin layer chromatogram. In addition, a single peak of formaldehyde-DNPH appeared at retention time of 4 minutes on the HPLC chromatogram (Fig. 2a).

2.6. Determination of Formaldehyde in Air and Leachate

Formaldehyde was analyzed according to the U. S. Environmental Protection Agency Method 8315A (US EPA 1996). A Waters 2695 high-performance liquid chromatography (HPLC) system with UV-Vis detector (Waters 996, Waters Corporation, Milford, MA) was used and chromatogram was recorded at 370 nm. The analytical column was a 3.9 × 150 mm HPLC column (Nova-Pac® C18 60Å 4 μm, Waters Corporation, Milford, MA). HPLC conditions were as follows: 40/60 acetonitrile/water (v/v), hold for 1 min; 40/60 acetonitrile/water to 100 percent acetonitrile in 3 min; 100 percent acetonitrile for 10 min; flow rate: 1.0 mℓ/min; injection volume: 20 μℓ.

Formaldehyde in air was derivatized on the DNPH cartridge then eluted with 1 mℓ of acetonitrile and injected into the HPLC-UV system. Leachate (1 mℓ) was derivatized by adding 0.5 mℓ of DNPH solution (100 mg/100 mℓ in 0.1N HCl) and mixing for 20 minutes. Derivatized formaldehyde was extracted with 1 mℓ of toluene, evaporated to dryness under nitrogen stream, dissolved in 1 mℓ acetonitrile and injected into the HPLC-UV system.

Formaldehyde concentrations in air and leachate were determined from a formaldehyde-DNPH standard curve. The formaldehyde concentration in the leachate was converted to total milligrams formaldehyde.

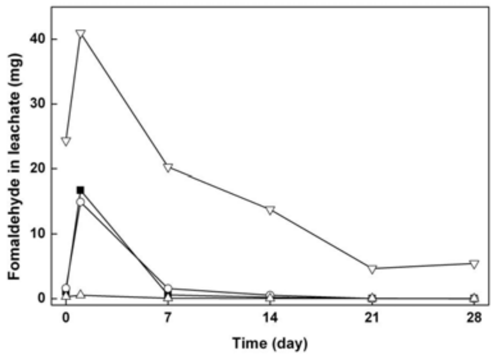

Fig. 3. Formaldehyde released in leachate from ground MDF in soil (■), cut pieces of MDF in soil (○), soil only (△), and ground MDF only (▽) buried in a simulated landfill for 28 days.

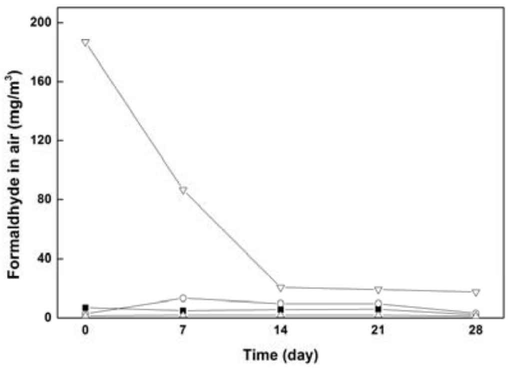

Fig. 4. Formaldehyde released in air from ground MDF in soil (■), cut pieces of MDF in soil (○), soil only (△), and ground MDF only (▽) buried in a simulated landfill for 28 days.

3. RESULTS and DISCUSSIONS

All opened containers were stored in an incubator set at 34℃ which represents an average summer temperature in the southern USA. Therefore, our data is not representing all the circumstances. Also, since a typical landfill contains mixed municipal solid waste, determination of formaldehyde released from only wood waste containing UF resin from a landfill is impracticable. An alternative plan for pursuing the objective of a study was to use simulated landfill constructed of alternating layers of soil and MDF. A plastic container with clamp lid and Tedlar bag containing nitrogen were used to avoid the influence of naturally occurring formaldehyde in the air. The simulated landfills successfully generated not only sufficient amount of leachate by adding water weekly, but also facilitated the collection of air. Our experimental design however did not fully represent a typical landfill and all environmental factors, but was successful in determining formaldehyde concentrations in air and leachate. Moreover, the amount of formaldehyde in air or leachate represented not accumulated amount of that. For better understanding or tracking of formaldehyde migration from MDF, total amount of formaldehyde in MDF may be helpful, but it was not possible to know in our experimental design because we did not make MDF sample.

3.1. Formaldehyde Emission in Air

Fig. 2c shows a typical chromatogram after DNPH derivatizing of air sample. Unknown aldehydes as well as formaldehyde were also detected in air samples compared to in leachate (Fig. 2b). A higher concentration of formaldehyde was released from the MDF only (treatment 4) compared to the treatments containing MDF and soil or soil only. The majority of free formaldehyde was emitted in the MDF only treatment during the first 14 days due to no interference by soil and high contact area with air. After 14 days, formaldehyde emissions were observed in constant level, but higher levels than other treatments (Fig. 4).

To better visualize the difference in formaldehyde concentration between different sizes of MDF in soil and the soil only, results from the MDF only treatment were excluded on Fig. 5. On day 0, approx-

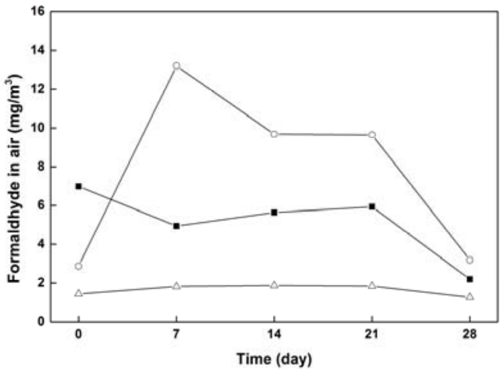

Fig. 5. Formaldehyde released in air from ground MDF in soil (■), cut pieces of MDF in soil (○), and soil only (△) buried in a simulated landfill for 28 days.

imately 2.5 times more formaldehyde was emitted from ground MDF in soil than in cut pieces of MDF in soil. However, in the second sampling time, the opposite occurred: approximately 3.5 times more formaldehyde was released in the cut pieces of MDF in soil than from ground MDF in soil. On day 21, approximately 1.5 times more formaldehyde was emitted from cut pieces of MDF in soil compared to ground MDF in soil. By the end of the study, the formaldehyde released from ground MDF or cut pieces MDF in soil were between 1.0 - 3.0 ppm. Formaldehyde emissions from soil only ranged between 1.0 - 1.5 ppm over the study.

There were no significant differences in formaldehyde air emission between ground MDF in soil and cut pieces of MDF in soil which indicates different MDF sizes, ground and cut pieces, did not affect the average formaldehyde air emission. However, there was significantly less formaldehyde emitted from soil amended MDF treatments than MDF only treatments. These results indicated that formaldehyde emission in air was greatly restricted by the soil.

Air emission of formaldehyde, however, was detected from all treatments at the end of the study (28 days) which may indicate formaldehyde is emitted slowly overtime from MDF disposed in soil or MDF only. The concentration of formaldehyde in air was detected at levels causing health issues under these experimental conditions.

3.2. Formaldehyde in Leachates

The formaldehyde-DNPH derivative and un-reacted DNPH were detected at retention times of 4 and 2.5 minutes, respectively (Fig. 2b). Other aldehyde-DNPH derivatives were also detected on the chromatogram. On day 1, the leachate from MDF only was highest in formaldehyde compared to MDF in soil (treatments 1 and 2) while the soil only contained the lowest amount of formaldehyde in the leachate (Fig. 3). The amount of formaldehyde significantly decreased by 50 percent in the ground MDF only treatment by day 7 compared to between 90 - 96 percent in the ground or cut pieces of MDF in soil 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 (day 28), the formaldehyde in the leachate was reduced by 99 percent in MDF in soil (treatments 1 and 2) and by 78 percent in the MDF only leachate. However, formaldehyde was still detected in the MDF only leachate.

Ground MDF or cut pieces of MDF in soil were not significantly different in formaldehyde released indicating that size (ground and cut pieces) of MDF did not affect formaldehyde amounts in leachate. However, there were significant differences in the amount of formaldehyde in leachates from ground MDF only (treatment 3) and MDF in soil (treatments 1 and 2) or soil only (treatment 4).

Overall, the formaldehyde reduction in leachate from treatments containing MDF and soil increased to 90 - 96 percent at day 14 and increased to 99 percent at the end of the study. From these results, nearly all of the free formaldehyde at this time was removed from the MDF in 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 microorganisms or binding to the soil. Moreover, soil type may influence the formaldehyde that is released into the air and leachate from the disposed MDF due to characteristics of soil such as, density, porosity and drainage.

4. CONCLUSION

In the treatments containing MDF and soil, the formaldehyde released into the air and leachate decreased to the formaldehyde level in soil only treatment by 28 days. However, in the treatment containing only MDF, formaldehyde was continuously released into the air and leachate. We conclude that formaldehyde was either trapped by soil, decomposed by soil microorganism or converted to other compounds. Therefore, soil reduces formaldehyde released from MDF disposed in soil. Disposing MDF in landfills may be a possible remediation method for wood waste containing formaldehyde-based resins. Based on formaldehyde release data, waste MDF may be used as mulch or compost after 1 month ground-up.

Future studies should include different soil types and soil chemistry in order to better understand the effect of formaldehyde concentration in the leachate and air. Additional experiments will be needed such as formaldehyde migration, reduction, and degradation in soil over time to address formaldehyde's lifetime in soil.

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