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Decomposition and carbon storage of selected paper products in laboratory-scale landfills

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HIGHLIGHTS

- Decomposition of major paper products measured under simulated landfill conditions
- Varied decomposition behaviors across paper types governed by pulp types
- A copy paper made from eucalyptus exhibited inhibited decomposition.

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ABSTRACT

The objective of this study was to measure the anaerobic biodegradation of different types of paper products in laboratory-scale landfill reactors. The study included (a) measurement of the loss of cellulose, hemicellulose, organic carbon, and (b) measurement of the methane yields for each paper product. The test materials included two samples each of newsprint (NP), copy paper (CP), and magazine paper (MG), and one sample of diaper (DP). The methane yields, carbon storage factors and the extent of cellulose and hemicellulose decomposition all consistently show that papers made from mechanical pulps (e.g., NPs) are less degradable than those made from chemical pulps where essentially all lignin was chemically removed (e.g., CPs). The diaper, which is not only made from chemical pulp but also contains some gel and plastic, exhibited limited biodegradability. The extent of biogenic carbon conversion varied from 21 to 96% among papers, which contrasts with the uniform assumption of 50% by the Intergovernmental Panel on Climate Change (IPCC) for all degradable materials discarded in landfills. Biochemical methane potential tests also showed that the solids to liquid ratio used in the test can influence the results.

1. Introduction

An estimated 64 million metric tons of paper and paperboard was generated in the U.S. municipal solid waste (MSW) in 2011 (U.S. EPA, 2013). Landfills remain the primary disposal method for MSW in the U.S., although other alternatives (e.g., recycling, combustion) are available in parts of the country. The disposal of paper products in landfills also occurs in other countries that rely heavily on landfills for MSW disposal. For example, Ximenes et al. (2008) reported that approximately two million metric tons of paper products were placed in Australian landfills each year.

Papers are made from fibers that are primarily comprised of cellulose, hemicellulose, and lignin, all of which contain biogenic carbon. The major products of the anaerobic decomposition of cellulose and hemicellulose in landfills are carbon dioxide and methane, a potent greenhouse gas (GHG). Some of the carbon dioxide and methane are released as a fugitive emission because they are either produced prior to the installation of a gas collection and control system, or gas collection is not 100% efficient. On the other hand, a significant portion of the biogenic carbon does not decompose, and therefore represents a source of carbon storage in landfills (Barlaz, 1998; Eleazer et al., 1997; Wang et al., 2011).

Information on the biodegradability of paper products in landfills is important in estimating methane emissions and carbon

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storage in landfills. For example, the Intergovernmental Panel on Climate Change (IPCC) has not only adopted a default decomposition factor (i.e., DOC_f — fraction of degradable organic carbon dissimilated) of 50% for all degradable materials placed in landfills (including paper) for inventory purposes (IPCC, 2006), but also allows reporting of methane emissions based on well documented country specific data. Biodegradability data are also used to support life cycle assessments of GHG impacts of consumer paper products (e.g., Dahlbo et al., 2005; Villanueva and Wenzel, 2007) and to estimate the effects of changes in MSW composition (due to the increased recycling and reuse) on changes in methane production and emissions from landfills (e.g., Levis et al., 2014).

Limited studies have attempted to measure the decomposition and carbon storage of different paper products under simulated or field landfill conditions. In simulated landfill reactors operated to maximize decomposition, Eleazer et al. (1997) reported methane yields and carbon storage factors of 74.3, 84.4 and 217.3 mL CH_4 g^{-1}, and 0.42, 0.27 and 0.05 g carbon stored g^{-1} initial dry material for newsprint, coated paper and copy paper, respectively. The higher lignin content in the newsprint, which is generally made from mechanical pulp, resulted in the lowest methane yield and highest carbon storage. Greater decomposition of copy paper with lower lignin content relative to newsprint has also been reported in field studies (Baldwin and Stinson, 1998; Wang et al., 2013). This is because lignin is recalcitrant under anaerobic conditions that exist in landfills and therefore limits the bioavailability of cellulose and hemicellulose, which are structurally associated with lignin (Colberg and Young, 1985; Stinson and Ham, 1995; De la Cruz et al., 2014).

The chemical composition of paper products has changed over time as more recycled fibers and inorganic materials are now more extensively used in the manufacturing process compared to the 1990s. For example, the ash content of copy papers has increased from 1.4% (Eleazer et al., 1997), 7.3% (Owens and Chynoweth, 1993), up to 12% (Wu et al., 2001) and 17% (Kong, 2008). This is consistent with the addition of inorganic fillers like calcium carbonate and clay into copy paper to improve printing properties (Chen et al., 2012). The objective of this study was to measure the anaerobic biodegradation of different types of paper products in laboratory-scale landfill reactors. This included measurement of methane yields and carbon storage factors (CSFs) for each paper product as well as the loss of cellulose, hemicellulose and organic carbon.

2. Materials and methods

2.1. Experimental approach

The anaerobic biodegradability and carbon storage of newsprint (NP), copy paper (CP), magazine (MG) and diaper (DP) were measured in 8-L reactors incubated under laboratory conditions designed to achieve the maximum decomposition in the minimum time. The test materials were selected to represent the major paper types in municipal waste and the range of expected biodegradabilities, including two samples each of newsprint (NP 1 and NP 2), copy paper (CP 1 and CP 2), and magazines (MG 1 and MG 2), and one diaper (DP) sample (termed Series A). As described with the Results, a second series of tests (termed Series B) was conducted with newsprint (NP 1 and NP 2) and copy paper (CP 1 and CP 2) to explore the consistency of initial results.

Incubation conditions were designed to maximize the rate and extent of decomposition, which included initial seeding with leachate from decomposing residential solid waste, leachate neutralization and recirculation, the periodic addition of nitrogen (N) and phosphorus (P) to maintain these nutrients above 100 mg NH_3–N L^{-1} and 5 mg PO_4–P L^{-1}, respectively, and incubation in a room maintained at about 37 °C. All reactors were monitored until they were no longer producing measurable methane.

2.2. Materials

Copy paper represents a chemical pulp in which most of the lignin has been chemically removed while newsprint represents a mechanical pulp in which the lignin is still present. Magazines may be made from either chemical or mechanical pulp, or a combination of both.

Materials were supplied by a consortium of pulp and paper companies in Australia. Two samples of newsprint (NP) were provided by Norske Skog in Australia. NP 1 is a standard newsprint, comprising 60% mechanical softwood fiber from *Pinus radiata* (radiata pine), 20% recycled fiber, and 20% semi-chemical hardwood pulp that was made by a process known as cold caustic soak from mixed eucalyptus species. NP 2 is an enhanced brightness newsprint made from 60% bleached softwood (pine) mechanical pulp, 20% recycled fiber, and 20% bleached semi-chemical hardwood (eucalyptus) pulp. The recycled fiber may contain a mixture of both chemical and mechanical pulp and this could vary over time and with the particular paper mill. Based on initial results, three additional newsprint samples were provided (termed NP A, B and C) to further explore variability in newsprint composition and biodegradability. These newsprints were described as a mix of virgin softwood and hardwood as well as recycled fiber that could be a mix of chemical and mechanical pulps and would include both hardwood and softwood.

Two samples of copy paper (CP) were provided by Fuji Xerox of Australia. CP 1 was manufactured from acacia fiber in Indonesia while CP 2 was made from eucalyptus fiber which is now the primary source of copy paper fiber in Asia. Two types of magazines (MG) were provided by Australian Paper. MG 1 is a chemical pulp while MG 2 contains mechanical fiber. Finally, diapers and a sample of the pure gel material contained therein were provided by Kimberly Clark Corporation. Diapers were comprised of approximately 60% pulp that originated from softwoods.

All materials except for the diapers were reduced in size using a paper shredder prior to use. In the case of diapers, one reactor was initiated with diapers that were cut into 3 cm by 2 cm strips and two additional reactors were filled with whole diapers that were folded to simulate their geometry as they would be disposed in a landfill. Unused diapers were tested to measure the decomposition attributable to the diaper material only.

2.3. Reactor construction, filling, and operation

The procedures for reactor construction, filling and operation have been described previously and are summarized here (Wang et al., 2011). Test materials were added to 8-L reactors in 7.5-cm layers and compacted using a specially fabricated tool designed to match the reactor diameter. Reactors were inoculated with leachate collected from a 300-L reactor that contained decomposing residential solid waste. Leachate was added to each layer and a total of 1 L was added to each reactor. Leachate, rather than decomposing refuse, was used so that the only significant source of biogenic carbon was the test substrate. Sufficient deionized (DI) water was added initially to ensure the generation of about 500 mL liquid for recirculation. DI water was also added periodically during the incubation period to maintain the recirculation liquid volume of 500 mL due to sample removal. In the Series B reactors, leachate was re-added to all reactors on Day 51 to provide additional inoculation. Gas was collected using a flex-foil gas bag (SKC Corp., Houston, TX) and leachate was collected in an intravenous bag (Baxter Healthcare, Deerfield, IL).

Reactor leachate was neutralized and recirculated as necessary throughout the incubation period. The concentrations of N and P were measured approximately monthly and adjusted to the aforementioned target concentrations with NH_4Cl and KH_2PO_4 as necessary. The pH and chemical oxygen demand (COD) were also measured weekly to bi-monthly throughout the incubation period.