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The role of biodegradation in limiting the accumulation of petroleum hydrocarbons in raingarden soils

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ABSTRACT

Previous studies have indicated that raingardens are effective at removing petroleum hydrocarbons from stormwater. There are concerns, however, that petroleum hydrocarbons could accumulate in raingarden soil, potentially resulting in liability for the site owner. In this work, 75 soil samples were collected from 58 raingardens and 4 upland (i.e., control) sites in the Minneapolis, Minnesota area, representing a range of raingarden ages and catchment land uses. Total petroleum hydrocarbon (TPH) concentrations in the samples were quantified, as were 16S rRNA genes for Bacteria and two functional genes that encode for enzymes used in the degradation of petroleum hydrocarbons. TPH levels in all of the raingarden soil samples were low (<3 μg/kg) and not significantly different from one another. The TPH concentration in raingarden soil samples was, however, significantly greater ($p \leq 0.002$) than TPH levels in upland sites. In addition, the number of copies of Bacteria 16S rRNA genes and functional genes were greater in the raingardens planted with deeply-rooted natives and cultivars than in raingardens containing simply turf grass or mulch ($p < 0.036$), suggesting that planted raingardens may be better able to assimilate TPH inputs. The ability of microorganisms present in the soil samples to degrade a representative petroleum hydrocarbon (naphthalene) was also investigated in batch experiments. A sub-set of the field sites was selected for re-sampling, and all soil samples tested ($n = 8$) were able to mineralize naphthalene. In these experiments the initial mineralization rate correlated with the number of copies of Bacteria 16S rRNA genes present.

1. Introduction

Pollution from urban stormwater runoff is a significant source of surface water impairment and presents one of the greatest water quality challenges of the coming century (National Research Council, 2008; U.S. EPA., 2000; U.S. EPA., 2005; Wang et al., 2000; Davis, 2005). Common pollutants detected in stormwater runoff include nutrients such as phosphorus and nitrogen, heavy metals, pathogens, and petroleum hydrocarbons (Davis et al., 2009). Petroleum hydrocarbons are washed from impervious surfaces such as parking lots and streets by rainfall, with reported concentrations in stormwater ranging from 0.2 to 277 mg/L (Wu et al., 1998; Kim et al., 2007, 2005; Zanoni, 1986; James et al., 2010). The petroleum hydrocarbons on such surfaces often result from leaks or spills of fuel, oils, and greases that are associated with motor vehicle use (Davis and McCuen, 2005). The impervious surfaces themselves can also serve as sources of petroleum hydrocarbons, as coal tar-based pavement seal coats are believed to be a significant source of polycyclic aromatic

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hydrocarbons (PAHs) to the environment (Yang et al., 2010; Van Metre et al., 2009; Mahler et al., 2005; Van Metre et al., 2000; Watts et al., 2010). Petroleum hydrocarbons are of concern because some compounds under this rather broad class of chemicals are toxic to aquatic life and some are known or suspected human carcinogens (Fent, 2003; Mastrangelo et al., 1996). Unfortunately, little research has been conducted on the removal of petroleum hydrocarbons from stormwater by best management practices (BMPs) and the ultimate fate of the removed hydrocarbons.

Increasingly, infiltration-based stormwater BMPs, including vegetated swales, infiltration trenches, and raingardens, are being implemented to reduce total runoff volume and ameliorate the environmental effects of stormwater through a variety of physical, chemical, and biological mechanisms (Davis et al., 2009; Li and Davis, 2009). Raingardens, also known as bioinfiltration or bioretention practices, are shallow vegetated depressions with an engineered soil media to which stormwater from impervious surfaces is directed for infiltration (Davis and McCuen, 2005; Dietz, 2007). In particular, raingardens have grown in popularity throughout the United States as a component of low impact development (LID). Because stormwater runoff contains petroleum hydrocarbons and other contaminants, it is important to determine the fate of these contaminants in raingardens.

Few studies have explicitly examined petroleum hydrocarbon removal in raingardens. Hsieh and Davis (2005) observed greater than 96% decrease of oils and grease in laboratory-scale raingardens and nearly 100% in full-scale systems. Hong et al. (2006) observed approximately 90% decrease of petroleum hydrocarbons in laboratory columns. DiBlasi et al. (2009) examined PAH removal at a bioretention field site and reported an average 87% mass load reduction. Based on the available evidence, bioretention appears to be successful in removing hydrocarbons from infiltrated stormwater (Davis et al., 2009; Weiss et al., 2008). Nonetheless, we are unaware of any research performed to investigate the ultimate fate of hydrocarbons in raingardens. Removal is presumed to be a combination of sorption and biodegradation (Hong et al., 2006), but these two mechanisms and their relative importance in raingardens have not been adequately explored.

Biodegradation of petroleum hydrocarbons is an oxidative process initiated by enzymes that can result in hydrocarbon mineralization; biodegradation has been observed in a wide array of environments, including under both aerobic and anaerobic conditions (Haritash and Kaushik, 2009). The genes that encode for oxygenases or other relevant enzymes can be enumerated using techniques such as quantitative polymerase chain reaction (qPCR). Molecular methods have the advantage of being rapid and specific, and can be used to target specific functional genes such as those encoding for oxygenase enzymes. Although the mere presence of a specific functional gene does not indicate that the reaction catalyzed by that enzyme is actually occurring, such molecular methods appear to be useful for assessing bioremediation activity or potential. For example, qPCR was employed to evaluate the progress of monitored natural attenuation at gasoline-contaminated sites (Baldwin et al., 2008, 2009). qPCR, however, has not yet been used to assess contaminant biodegradation capacity in bioretention systems.

Although the removal of petroleum hydrocarbons from runoff prior to discharge to a receiving water is beneficial, the potential for accumulation of pollutants in raingarden soils is a significant concern (Davis et al., 2003). For example, PAH accumulation in the sediments of stormwater retention ponds has been reported (Van Metre et al., 2009; Van Metre et al., 2000; Kamalakkannan et al., 2004), and can lead to prohibitively high disposal costs for the dredging spoils. If such “toxic depots” are created in raingardens, there is potential environmental liability to the site owner, which could decrease the rate of implementation of these beneficial LID technologies. Therefore, it is important to establish if petroleum hydrocarbons accumulate to unsafe levels in raingarden soils.

Biodegradation of petroleum hydrocarbons occurs readily under aerobic conditions when other criteria such as pH, temperature, and nutrient levels do not limit microbial growth (Mohn and Stewart, 2000; Zhou and Crawford, 1995). Thus, it appears that petroleum hydrocarbon degraders are ubiquitous in the environment (Haritash and Kaushik, 2009; Margesin et al., 2003). As aerobic conditions are typical in shallow soil environments, it is expected that petroleum hydrocarbons would be readily biodegradable in raingardens. Nonetheless, raingarden media is designed to promote rapid infiltration, resulting in short hydraulic residence times that may be insufficient to promote biodegradation. Furthermore, the organic matter in raingarden media from the addition of compost will likely limit the bioavailability of petroleum hydrocarbons. Raingardens commonly experience variable pollutant loading and inconsistent soil moisture conditions (Davis and McCuen, 2005), which could inhibit the accumulation of petroleum hydrocarbon degraders. Therefore, this research was performed to examine the concentration of petroleum hydrocarbons in raingarden soils at field sites, to evaluate the potential for petroleum hydrocarbon biodegradation in raingarden soils, and to investigate what factors influence petroleum hydrocarbon residual. Finally, qPCR was investigated as a rapid method to evaluate the biodegradation potential of bioretention soils.

2. Materials and methods

2.1. *Field sampling and site classification*

A total of 75 soil samples were collected from May to August, 2008 in the Minneapolis-St. Paul metropolitan area (Minnesota, USA); 71 of the samples were from raingarden sites and 4 additional upland samples served as controls. Fifty-eight raingardens were sampled, with one soil sample collected from 49 of the raingarden sites and multiple soil samples (up to 4) collected from 9 of the raingardens. Soil samples (approximately 10 g each) were collected just below the ground surface (or below the mulch layer, if present) at the raingarden entrance (e.g., curbcut) and/or near the lowest elevation of the raingarden. Samples were collected using sterilized scoops and were placed in autoclaved glass bottles. The raingarden sites were all infiltration-based urban stormwater best management practices and represented a wide array of sizes, vegetation types, catchment land uses, and catchment areas. In addition to the samples collected from