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Research article

Geomembrane applications for controlling diffusive migration of petroleum hydrocarbons in cold region environments

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ABSTRACT

Laboratory permeation tests examine the migration of aromatic hydrocarbons (benzene, toluene, ethylbenzene and xylenes (BTEX)) at 2, 7 and 14 °C through three different types of geomembrane (high density polyethylene (HDPE), linear low density polyethylene (LLDPE) and polyvinyl chloride (PVC)). Tests on both virgin and exhumed field samples provide permeation parameters (partitioning (S_{gf}), diffusion (D_g), and permeation (P_g) coefficients) for the three geomembranes. These results are combined with published values for the same geomembranes at 23 °C to establish an Arrhenius relationship that can be used to estimate diffusion parameters at temperatures other than those for which tests were conducted. Tests on an HDPE geomembrane sample exhumed after 3 years from a landfill site in the Canadian Arctic showed no significant difference in diffusion characteristics compared to an otherwise similar unaged and unexposed HDPE geomembrane. Contaminant transport modeling for benzene through HDPE, LLPDE and PVC in a simulated landfill cover show that for the conditions examined the presence of any of the three geomembranes below the 2 m thick soil cover substantially reduced the contaminant flux compared to the soils alone for realistic degrees of saturation in the cover soil. For these same realistic cold climate cases, of the three geomembranes examined, the HDPE geomembrane was the most effective at controlling the contaminant flux out of the landfill. An increase in soil cover and liner temperature by 2 °C (from potential climate change effects) above those currently measured at an Arctic landfill showed an increase in contaminant transport through the cover system for all geomembranes due to the increase surface temperature (especially in the summer months). Modeling of the addition of an extra 0.5 m of soil cover, as a mitigation measure for the effects of climate change, indicates that the main benefit of adding this unsaturated soil was to reduce the geomembrane temperature and that this did reduce the magnitude of the increase in contaminant transport.

1. Introduction

Hydrocarbon fuels continue to be the main source of energy used in the Arctic, Antarctic and other cold climatic regions. Poor handling practices and the large volume of hydrocarbons required for operations has led to the contamination of these once pristine environments. A number of innovative approaches have been developed to remediate hydrocarbon-contaminated soil, water and source plumes in these areas including bioremediation, landfarming, landfills, containment barriers, biopiles and permeable treatment barriers (Van Stempvoort and Biggar, 2008; Snape et al., 2008; Paudyn et al., 2008; Kalinovich et al., 2008, 2012; Sancartier et al., 2009; Mumford et al., 2013, 2014; McWatters and Rowe, 2015).

As geoenvironmental applications evolve in these cold regions, the use of geosynthetic materials becomes more significant, especially geomembranes. The primary function of a geomembrane, whether existing as a single liner or incorporated into a composite liner barrier system, is to minimize the diffusive and advective migration of contaminants in either a vapour or liquid state, through base liners and final covers and into the surrounding environment (Rowe, 2005; Shackelford, 2014). This is important in the Polar Regions where the ecosystems are more sensitive to

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contamination and the impact of human activities (Snape et al., 2001; Poland et al., 2003).

In cold regions, geomembranes have been used as protective layers surrounding diesel tanks or supplies, landfill base liners and covers, in temporary containment barriers and as collection pond liners for permeable reactive barriers, treatment walls or in mining applications (Richards and Foster, 1991; Bathurst et al., 2006; Kalinovich et al., 2008, 2012; Mumford et al., 2013; McWatters et al., 2016; Hosney and Rowe, 2014). Regulated land farms often require a protective base layer (Filler et al., 2006), which in some cases can be provided by a geomembrane. Geomembranes were employed and studied for the first time in an Antarctic environment at Casey Station, as a layer in the composite liner barrier system of biopiles (or contained treatment cells) used to remediate hydrocarbon-contaminated soil (McWatters and Rowe, 2015). There is also potential for geomembranes to be used as vapour barriers to impede intrusion of volatiles into buildings especially when cold outdoor temperatures are in contrast to the elevated indoor temperatures of buildings (Garetto et al., 2012).

Geomembranes have been used for a variety of applications during remediation of a contaminated former military base on the summit of Resolution Island, Nunavut (61° 350N, 60° 400W) in the Canadian Arctic. This site was part of the Distant Early Warning Line, an integrated system of more than 42 radar and communication sites that stretched 4800 km across the Arctic, established in the 1950s. This larger site was heavily contaminated with polychlorinated biphenyls (PCBs), hydrocarbons and metal during and after its years of operation (Poland et al., 2003). Remediation of the site proceeded in early 1990s until 2006; thereafter a long-term monitoring program began (Kalinovich et al., 2012). During remediation activities, geomembranes were used in the base barrier system of four permeable reactive barriers (PRBs) designed to filter and treat PCB and hydrocarbon surface water from the spring thaw (Kalinovich et al., 2008, 2012; Paudyn et al., 2008). Geomembranes have also been used in the landfill, constructed onsite to contain contaminated soils (ASU, 2005). In total 134 landfills were built at 21 of the 42 Arctic Dew Line remediated sties and some of these were designed with geomembranes in the barrier system (Contenta, 2012). The Resolution Island landfill, like others designed for the polar tundra climate of the Canadian Arctic, uses the permafrost as a means of containment and as freezeback occurs, the landfill itself becomes a part of the permafrost layer below the active soil layer (Magee and Rice, 2002).

If complete freezing is achieved and maintained in perpetuity then, if saturated, the frozen ground would act as the primary barrier to outward migration of hydrocarbons. If unsaturated (dry or mixed saturation) the frozen ground would not be a good barrier to outward migration of hydrocarbons. Therefore, in order to establish potential contaminant migration, it is important to consider the length of time for freezeback to occur, the short and long-term thermal ground temperatures in the landfill and the depth of the annual thaw of the active layer in the material. Also important is the potential for changing depths of the active layer with increasing temperatures in the Canadian Arctic as a result of the changing climate (Schuur et al., 2013).

A geomembrane plays a significant role in controlling contaminant migration (a) before complete freeze of the waste and adjacent soils; (b) where there is potential for lateral movement of contaminants in the active layer (Rowe et al., 2007; Barnes and Biggar, 2008); (c) when vapour migration occurs in unsaturated soil (even if frozen); or (d) in the vapour or liquid-phase when there has been discontinuous permafrost and ice formation in the active layer that created fissures through which contaminants could escape (Barnes and Biggar, 2008).

Permafrost soils in cold climates can have a low capacity for sorption of hydrocarbons as well as low capacity for biological decay (Ping et al., 2015). The lower natural attenuation rates in these regions (Snape et al., 2008) can allow a greater aerial distribution of contaminants than would be expected in the soils found in more temperate areas (Mohn and Stewart, 2000). While the summer months bring slightly warmer temperatures, which may potentially increase bioremediation (Mohn and Stewart, 2000), they also increase the risk of mobilizing contaminants as the ground thaws (Snape et al., 2002; Rowe, 2006).

There has been relatively little research into the performance of barrier systems in cold climates (Rowe et al., 2007) and even less specifically on geomembranes in cold climates (Rowe et al., 2010), despite the widespread application of engineered landfill in these regions. Some studies have looked at physical and chemical changes in the polymer structure with exposure to cold temperatures (Bathurst et al., 2006; Rimal et al., 2007; Rowe et al., 2007, 2010), and freeze-thaw cycles (Hsuan et al., 1994, 1997, 2013; Comer et al., 1995). Changes in the diffusive parameters resulting from freeze-thaw cycling have not been studied. In the case of the freezeback landfill design, in the period before complete freezing occurs (permafrost aggradation), the performance of the geomembrane during this stage is important. It is also important should climate change affect the long-term thermal regime of these freezeback landfills by increasing the temperatures in the active soil layers.

Migration of contaminants through a composite liner barrier systems occurs by either advection or diffusion. A well-constructed system incorporating a geomembrane (with minimal holes) can minimize or completely remove the advective transport of contaminants. Under these circumstances, diffusion becomes the dominant mode of transport for contaminants (Rowe et al., 2004; Rowe, 1998, 2005). Diffusive transport of contaminants (such as hydrocarbons) through geomembranes is characterized by the partitioning (S_{gf}), diffusion (D_g), and permeation (P_g) coefficients (e.g. Sangam and Rowe, 2001).

The geomembranes used as liners in barrier systems are manufactured from different polymers. Common types include high-density polyethylene (HDPE), linear low density polyethylene (LLDPE) and polyvinyl chloride (PVC). Polyethylene is an excellent barrier to water and ions but is permeable to hydrocarbons (Sangam and Rowe, 2001; Feldman, 2002). PVC offers some resistance to chemicals and its greater flexibility is advantageous for smaller geomembrane applications (Ortego et al., 1995). The contaminant mass flux can be important for evaluating the performance of a landfill cover barrier system as this quantifies the diffusive transport of volatile contaminants moving through the intact liner system.

Several investigators have examined the diffusion of contaminants through HDPE geomembranes at 20–25 °C (Park and Nibras, 1993; Xiao et al., 1997b; Aminabhavi and Naik, 1998, 1999; Sangam and Rowe, 2001, 2002, 2005: Joo et al., 2004, 2005; Chao et al., 2007; Islam and Rowe, 2009). Park and Nibras (1993) and Aminabhavi and Naik 1998 examined diffusion of contaminants through LLDPE while Xiao et al. (1997a) and Nefso and Burns (2007) examined diffusion through PVC all with a focus on room temperature (or above) and thus there is a need for data at lower temperatures relevant to specific applications in cold climates.

Benzene, toluene, ethylbenzene and xylene (BTEX) are aromatic hydrocarbons commonly found at low concentrations in landfill gas and leachate (e.g., USEPA, 2003; Rowe et al., 2004) and in higher concentrations in hydrocarbon-contaminated soils or spill sites (Snape et al., 2008). Standard unleaded gasoline (petrol) blends can vary in amounts (w/w) of benzene (0.1–4.9%), toluene