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Nathalie Touze

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Performance of geosynthetics for environmental protection in landfills

N. Touze-Foltz

Irstea, Antony, France.

ABSTRACT: The objective of this Keynote Lecture is to give an updated overview of transfer properties of geosynthetic liner materials used in landfills and of parameters that impact them. Some innovative measurement techniques are detailed, especially as regards the measurement of advective flow rates for multicomponent geosynthetic clay liners (GCLs). The measurement of advective flow rates through geomembranes and multicomponent GCLs is first described. Then the effect of the mass per unit area of bentonite in GCLs, the nature of the bentonite in GCLs and of hydration without load on the hydraulic conductivity of GCLs are described. A brief presentation of pollutants of concern in landfills and especially of microorganic pollutants is given. The chemical compatibility of GCLs with leachate is discussed in the third section of this Keynote Lecture. Finally, the diffusion of organic and inorganic species in relation with the composition of leachate previously described is given in the last section of this Keynote Lecture.

1 INTRODUCTION

In recent years there have been many advances in the understanding of issues related to the use of geosynthetics such as geomembranes and geosynthetic clay liners (GCLs) as contaminant barriers.

The objective of this Keynote Lecture is to focus on the performance of geosynthetics for environmental protection in landfills. The geosynthetics under study will be those which function is to ensure lining, i.e. geomembranes and geosynthetic clay liners. Geomembranes are defined in the Recommended Descriptions of Geosynthetics Functions, Geosynthetics Terminology, Mathematical and Graphical Symbols of the International Geosynthetics Society as planar, relatively impermeable, polymeric (synthetic or natural) sheet used in civil engineering applications. Geosynthetic clay liners (GCLs) are defined as an assembled structure of geosynthetic materials and low hydraulic conductivity earth material (clay), in the form of a manufactured sheet, used in civil engineering applications. The EN ISO 10318 standard (AFNOR 2006) defines geosynthetic barriers, that can be polymeric, bituminous or clay geosynthetic barriers, depending on which component fulfils the barrier function. In the case of the clay geosynthetic barrier (GBR-C) the definition does not allow to

use GBR-C and GCL interchangeably as will be seen in the development of this Keynote Lecture. Indeed, multicomponent GCLs have been introduced to the market. The following definition proposals are currently being discussed in the ASTM D35 terminology task group and might be added in future in the ASTM terminology standard D4439 (von Maubeuge et al. 2011). A multicomponent GCL is a GCL with an attached film, coating, or membrane decreasing the hydraulic conductivity or protecting the clay core or both. An adhered GCL is a GCL product in which the clay component is bonded to a film or membrane by adhesion. A coated GCL is a GCL product with at least one layer of a synthetic substance applied to the GCL as a fluid and allowed to solidify (von Maubeuge et al. 2011). In this case as will be illustrated in Section 2.2 it is possible that the coating or attached film ensures the liner function, much more than the clay, which is in contradiction with the definitions of GBR-Cs. In the following it will thus be preferably referred to geomembranes and geosynthetic clay liners. The wording multicomponent GCL will systematically be used in the following when the GCL contains a coating or an attached film for a clear distinction of the various materials.

A barrier material is intending to control contaminant transport and ensure long term environmental impact. To ensure this, ways of quantifying

transfers through liner materials were developed, some of which very recently.

This Keynote Lecture will address the quantification in the laboratory of transfer parameters for geomembranes and geosynthetic clay liners. The elementary transfer modes on which the Keynote Lecture will focus are advection, thus the transport of liquid due to a difference in hydraulic head between both sides of a liner material, and diffusion, corresponding to a transfer linked with the existence of two different concentrations of a given contaminant on both sides of a liner material. No attempt will be made here to quantify the performance of barrier systems at field scale taking into account the combined effect of advection and diffusion, as this aspect has been previously extensively addressed for example by Rowe (2005, 2007, 2012). Rather the objective here is to discuss the material properties of geomembranes and GCLs and the way they can be quantified, insisting on recent innovative developments and resulting recommendations for the use of those materials in landfills.

The quantification of advective transfers in geomembranes, and geosynthetic clay liners, including multicomponent GCLs, will first be presented. Some of the properties impacting the hydraulic behaviour of GCLs will be highlighted like the effect of the nature of the bentonite, of the mass per unit area of bentonite, and of the hydration process. The quantification of flow rates in multicomponent GCLs due to the existence of a hole in the coating or the attached film will also be presented and compared to the more classical case of a composite liner made of a geomembrane and a GCL.

An insight will then be given on the composition of leachate focusing mainly on microorganic pollutants.

The chemical compatibility of GCLs with leachate will then be addressed with the objective to give clues as regards the selection of a bentonite for a given application.

Finally, diffusive transfers through geomembranes and GCLs will be addressed both for organic and inorganic species.

It is believed that the elements included in this Keynote Lecture will bring information that will help to better understand the behaviour of geosynthetics used for lining applications and tools for the quantification of their transfer properties.

2 QUANTIFICATION OF ADVECTIVE TRANSFERS THROUGH LINER MATERIALS AND FACTORS IMPACTING THEIR HYDRAULIC PERFORMANCE

The objective of this section is to address the quantification of flow rates through geomembranes and

GCLs, including multicomponent GCLs. The flow rate measurement through geomembranes will first be discussed in Section 2.1 as it serves as a basis for the innovative quantification of flow rates through multicomponent GCLs that will be presented in Section 2.3. The quantification of the hydraulic conductivity of GCLs will be briefly discussed in Section 2.2 and the effect of the nature of the bentonite, of the confinement during hydration and of the mass per unit area of bentonite on the hydraulic performance will be addressed.

2.1 *Flow rate measurement through geomembranes*

Geomembranes are non porous media. It means that there is no void in the material, but only free spaces which size is in the range of solvent's molecule size. Transport in geomembranes does thus occur at the molecular level (Lambert & Touze-Foltz 2000). However gases and liquids can migrate through the intact geomembranes by an activated diffusion process, different from the liquid convection process occurring through the pores of porous soils (Barroso 2005). Different driving forces may cause diffusion: concentration, hydraulic or temperature gradients. It has also been shown that diffusion occurs even if there is no gradient: this phenomenon is called self-diffusion (Eloy-Giorni 1993).

An equipment was first developed in France (Eloy-Giorni 1993, Pelte 1993, Durin et al. 1998, Lambert & Touze-Foltz 2000) in order to quantify flow rates through geomembranes linked with the application of a hydraulic gradient.

This development gave rise to a French standard and afterwards to a European standard (EN 14150) for the measurement of the steady-state liquid flow through geomembranes. The test method and described apparatus allow the measurement of flows accurately down to $10^{-6} \text{ m}^3/\text{m}^2/\text{d}$.

The two-part cell described in EN 14150 (see Figure 1) is made of stainless steel as the cell shall resist oxidation during long-term immersion. In each part of the cell, a cavity allows to apply a hydraulic pressure. A porous disc placed in the downstream cavity prevents deformations of the geomembrane.

The cell shall be designed to clamp the specimen without any leaks. There is no tightening system necessary, as clamping between flat surfaces is usually sufficient. In the case of bituminous geomembranes, a bitumen rubber sealant can be used.

The minimum diameter of the measuring chambers is 0.2m. The cell is equipped with a liquid inlet on the upstream part and a liquid outlet on the downstream part and flushing valves on each part.

The volume measurements can be achieved using capillary tubes (Type A device) or pressure-volume controllers (Type B device). A synthesis of measurements performed with type B device are presented on Figure 2. This device allows the application of a constant pressure when measuring the volume. It consists of a cylinder in which a piston slides. A numerically controlled motor enables the application of the required pressure by moving the piston. A pressure sensor included in the system measures the pressure. The piston displacement corresponds to a variation of the volume of liquid. As can be noticed for virgin geomembranes most flow rates are lower than 10^{-6} m³/m²/d. With a type B device, at least three temperature transducers, placed on each pressure-volume controller and on the cell, are used. Temperature measurements are then used to correct volume variations.



Figure 1 . View of the stainless steel cell and pressure volume controllers of type B device for flow rate measurement in geomembranes

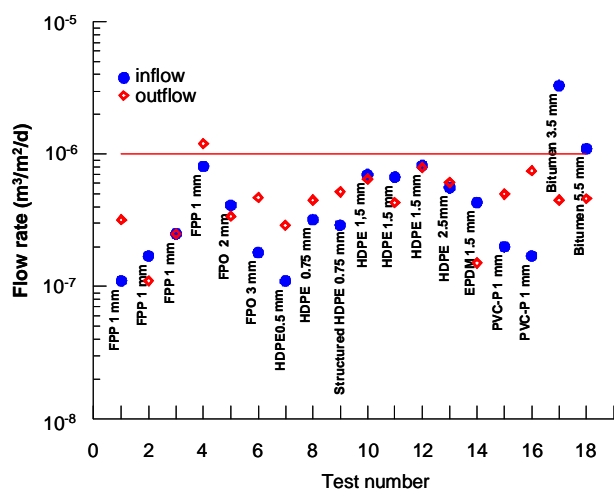


Figure 2. Synthesis of flow rate measurements through geomembranes according to EN 14150 with Type B device (from Touze-Foltz & Zanzinger 2009) (FPP: flexible polypropylene, EPDM: ethylene propylene diene monomer, HDPE: high density polyethylene, PVC-P: plasticized polyvinyl chloride, FPO: flexible polyolefine)

Validity of the measurement is examined by comparing upstream and downstream flow rate values. Theoretically, these values should be equal, but, in practice this is rarely the case for geomembranes.

For flow rates values greater than or equal to 10^{-6} m³/m²/d, upstream and downstream flow rates are considered as equal if the difference between them is less than 10 % of the measured flow rate on the upstream side.

In particular circumstances where testing according to the described test method indicates that values obtained for a geosynthetic barrier lies below the threshold of sensitivity of the test method then the value of liquid flow will be declared as being less than 10^{-6} m³/m²/day. Most values are below the threshold for the geomembranes tested.

The test device defined by EN 14150 has also been used to quantify the evolution of geomembranes after exposure (Touze-Foltz et al. 2010) for the case of bituminous geomembranes in hydraulic applications but it could be used for other geomembranes in the case of landfill applications provided that specimens are available for testing.

2.2 Measurement of the hydraulic conductivity of GCLs and factors influencing the hydraulic conductivity of GCLs on the short-term

2.2.1 Hydraulic conductivity measurement and synthesis of factors of influence

In the absence of a GM, the leakage through a clay liner is given by Darcy's law:

$$Q = Ak_L i \quad (1)$$

where Q = leakage through the liner (m³/s); A = area of liner under consideration (m²); k_L = hydraulic conductivity of the clay liner (m/s); and i = hydraulic gradient (-).

The hydraulic conductivity of GCLs is usually measured using ASTM D5887 (ASTM 2009), especially as far as CE marking is considered, or D5084 (ASTM 2010) that commonly involve a consolidation pressure of 35 kPa, a pressure difference across the specimen of 15 kPa and a permeant that is de-aired, deionized water (ASTM 2009), although de-aired tap water is also used by some manufacturers when using ASTM D5084 (Rowe 2012).

Alternatively, in France, rigid wall permeameters are used following NF P84-705 (AFNOR 2008) where variable loads from 10 to 200 kPa can be applied, jointly with hydraulic heads from 0.1 to 1.2m.

The short-term hydraulic conductivity, k_L , of a GCL will depend on (Rowe et al. 2004):

- The nature of the bentonite in the GCL (e.g., sodium or calcium); this point will be addressed in Section 2.2.2 of this Keynote Lecture,
- To some extent, the mass per unit area of bentonite; this point will be addressed in Section 2.2.3 of this Keynote Lecture,
- The method of manufacture of the GCL, especially in relation with the load applied to confine the bentonite during hydration, as will be discussed in Section 2.2.4 of this Keynote Lecture,
- The effective stress; this point will not be discussed here. The readers can refer for example to Petrov et al. (1997) for more information on this point,
- Bentonite migration down-slope in either a "dry" or hydrated state, and
- Lateral movement (thinning) of bentonite during and following hydration that would cause an uneven distribution of the bentonite in the GCL.

2.2.2 Influence of the nature of the bentonite in the GCL

Figure 3 shows the cationic composition of the bentonite in eight different GCLs representative of the European market as far as the use at the bottom of landfills is concerned, tested by Guyonnet et al. (2009). One has to observe that sodium or calcium for a bentonite means that sodium or calcium is the main cation present between the clay platelets but not the sole one. As natural sodium bentonites do not necessarily always perform better than calcium activated bentonites as will be discussed in Section 3, Guyonnet et al. (2009) suggested to group natural sodium and calcium activated bentonites under the wording sodium bentonites.

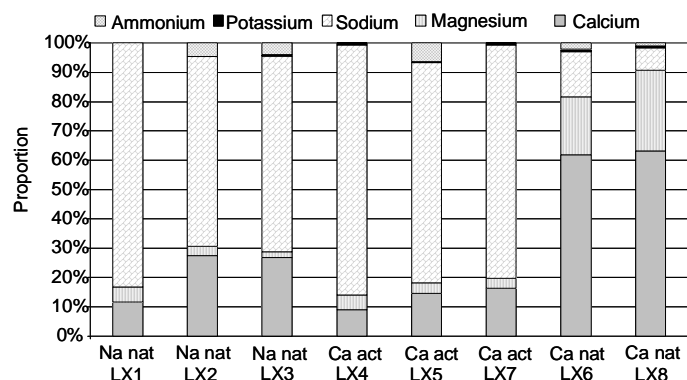


Figure 3. Main cation proportion in the various GCLs LX1 to LX8 tested by Guyonnet et al. (2009)

Figure 4 illustrates for those 8 GCLs the variation in hydraulic conductivity measured according to NF P84-705 (AFNOR 2008). LX1 to LX3 containing natural sodium bentonite and LX4, LX5 and LX7 containing calcium activated bentonites exhibit hydraulic conductivities lower than 3×10^{-11}

m/s under a 100 kPa confining stress. For the two GCLs containing calcium bentonite LX6 and LX8 the hydraulic conductivities under the same conditions are respectively 4.5×10^{-9} and 4.7×10^{-10} m/s.

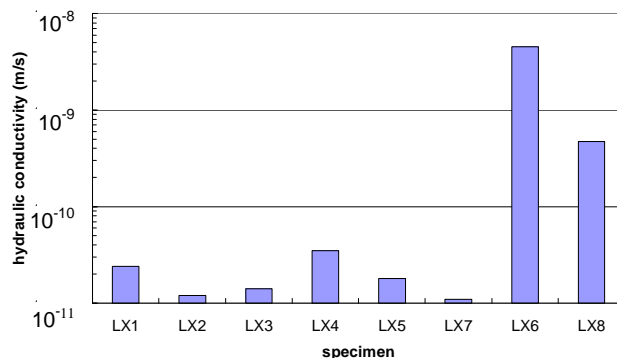


Figure 4. Hydraulic conductivity to NaCl 10^{-3} M of GCLs LX1 to LX8 from Guyonnet et al. (2009)

2.2.3 Effect of the mass per unit area of bentonite on flow through a GCL

The guide of recommendations for the evaluation of equivalency in passive barrier of municipal solid waste landfills in France (MEEDDAT 2009) specifies that when a GCL is used to reinforce a clay layer to ensure equivalence, the dry mass per unit area of bentonite in the GCL shall be 5 kg/m^2 . This requirement is consistent with the observation made by Bostwick et al. (2010) that GCL specimens with an uneven bentonite distribution (typically those with a low mass per unit area) can have areas of little to no bentonite content. A minimum mass per unit area is thus required which is 5 kg/m^2 for bottom landfill liners in France.

No confidence interval is given on this value. No indication is however given whether this value is an average value or a minimum mass per unit area for the whole surface of the GCL installed. The mass per unit area of bentonite in GCLs can be measured according to EN 14196, on five specimens (AFNOR, 2004). The standard indicates that the average value of the mass per unit area shall be given for the constitutive geosynthetics and for the whole GCL. It gives no indication regarding the mass per unit area of dry bentonite, which can be given as an average for each specimen.

The arising question is related to the interpretation or misinterpretation which can occur as the expression of the result is concerned, and its implication. Indeed, for a given GCL it is possible that the average mass per unit area is larger than 5 kg/m^2 but that the minimum mass per unit area on the roll is lower than 5 kg/m^2 . Shall the product then be rejected? Will a GCL with a dry mass per unit area equal to 5 kg/m^2 behave better or worse than a GCL for which the minimum mass per unit area of dry bentonite is 5 kg/m^2 ? In order to address this question a synthesis of measurements of

flow rates performed in the laboratory according to NF P84-705 (AFNOR 2008) was made for two different needle punched GCLs containing natural sodium bentonite, from two different manufacturers. GCL1 contains powdered bentonite while GCL2 contains granular bentonite. Results are reported on Figure 5 in terms of flow rate as a function of the mass per unit area of dry bentonite in each GCL specimen for a confining load equal to 10kPa. Similar results were observed for larger loads but with less data available. One can notice that even though variations in flow rates are small, a minimum flow rate is obtained for a dry mass per unit area of bentonite close to 5.5 kg/m². It would thus be better to interpret the recommendation of a mass per unit area equal to 5 kg/m² as a minimum mass per unit area of bentonite to ensure the smallest possible flow rate in the GCL.

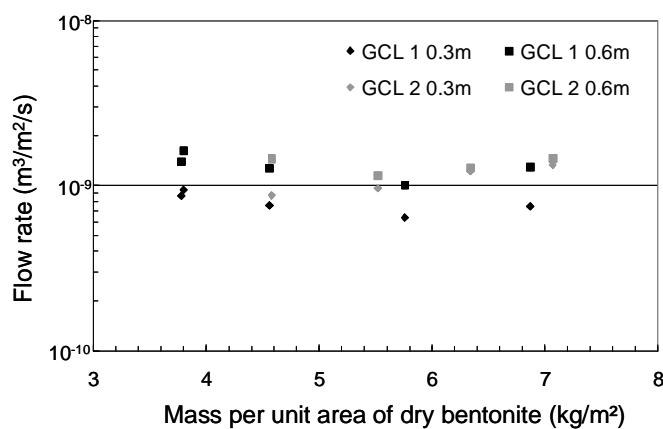


Figure 5. Evolution of the flow rate in two GCLs as a function of the dry mass per unit area of bentonite

2.2.4 Effect of the confinement during hydration on the hydraulic performance of GCLs

In order to perform properly GCLs have to be confined and hydrated. It is usually recommended that load be applied on top of the GCL immediately after installation. Indeed, Petrov et al. (1997) have shown that confining the GCL prior to hydration contributes to decrease the void ratio and thus to reduce the hydraulic conductivity. Nevertheless, there are a number of situations in which such a practice is not possible and GCLs may be left exposed to wetting without being confined. This can also arise when it is thought that a sufficient amount of water will not be supplied from the underlying soil to ensure hydration of the GCL.

Touze-Foltz et al. (2009) did study the effect of a 5 days hydration period, either due to immersion of a GCL or to a rainfall event for a GCL either installed vertically (see Figure 6) or horizontally (see Figure 7) on the swell of GCLs and on their hydraulic performance. Results were compared to the situation where the hydration would have taken place under a 20 kPa load. Three needle punched GCLs and two stitch bonded GCLs were studied.

The minimum mass per unit area of dry sodium bentonite in the GCLs was 5 kg/m² except for one of the stitch bonded GCLs (4.2 kg/m²). Flow rates measurements were performed according to NF P84-705 (AFNOR, 2008).



Figure 6. Hydration vertically in the heavy rainfalls situation.



Figure 7. Hydration horizontally in the heavy rainfalls situation.

Results obtained under a 20 kPa load as compared to the results obtained in the case the hydration took place under load showed a very little effect, by a factor 2 on the flow rate for needle punched GCLs. Results obtained for the stitch bonded GCLs were more variable depending on the mode of fabric, resulting in a very detrimental effect of the immersion for one of those GCLs.

Those results are consistent with previous results from Petrov et al. (1997) who have shown the beneficial impact of needle punching to restrict the swelling capabilities of GCLs subjected to low confining stresses and thus to significantly impact their hydraulic conductivity.

Those results resulted in the recommendation to confine stitch bonded GCLs immediately after installation in France for the use of GCL in tunnels and underground structures.

2.3 Flow rate measurement through intact multicomponent GCLs

Barral & Touze-Foltz (2012) developed a measurement technique based on two existing standards. The first one, NF P84-705 (AFNOR 2008) is dedicated to the measurement of flow rates through GCLs in rigid wall permeameters. The second one, EN 14150 (AFNOR 2006) is dedicated to the measurement of flow rates of water through geomembranes and was previously presented in Section 2.1. Basically, the testing cell from NF P 84-705 was used in combination with the measuring devices from EN 14150. The reason for this combination is that the test cell of EN 14150 is not suitable for accommodating GCLs specimens. Furthermore, the measuring device from NF P84-705 is not suitable for measuring flow rates as small as the ones crossing geomembranes or geofilms. This did thus result in the combination of testing devices from those two standards. Figure 8 gives an insight in the resulting testing apparatus.

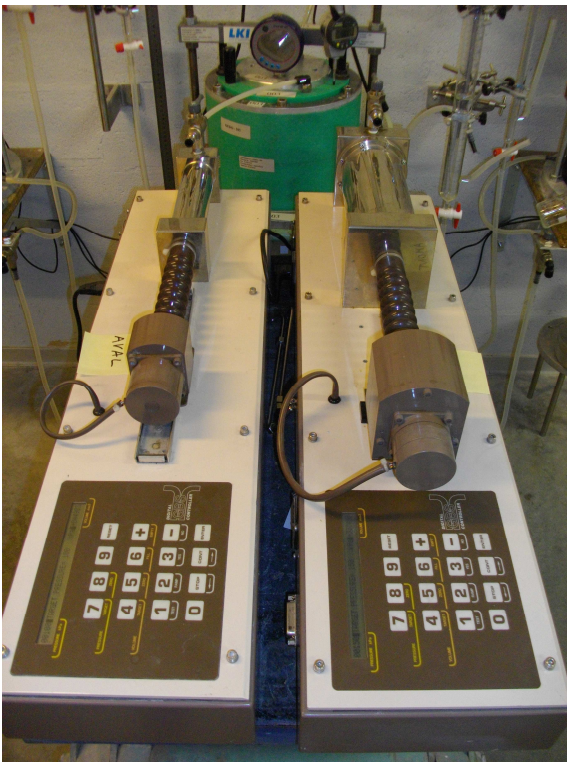


Figure 8. Experimental device for measurement of flow rates through multicomponent GCLs

Three different multicomponent GCLs were tested with this procedure, presented in Table 1. Figure 9 shows some of the results obtained. Multicomponent GCLs 1 and 2 contain an attached film 0.1mm or 0.2 mm thick whereas multicomponent GCL 3 is coated with a mass per unit area of

coating larger than 200 g/m² according to the manufacturer.

Multicomponent GCLs were hydrated under a 10 kPa confining load and with a hydraulic head equal to 0.01m. Water is applied on the side of the multicomponent GCL opposite to the coating or attached film. Once 95% of the absorption and the swell for an infinite time are obtained (see NF P84-705 for calculation details), the confining load and hydraulic head are increased.

In the test, the multicomponent GCLs were confined using a 170 kPa load. A pressure equal to 150 kPa was applied at the upstream side of the cell, while a 50 kPa pressure or larger was applied at the downstream side. Pressures equal to 150 kPa and 50 kPa are identical to the ones applied in EN 14150.

Table 1. Features of the multicomponent GCLs studied

Multicomponent GCL	Cover GTX	carrier GTX	Coating or film thickness (mm)	Mass per unit area of dry specimen (kg/m ²)
1	woven	nonwoven	≈0.1	4.41
2	nonwoven	woven	≈0.2	5.28
3	woven	nonwoven	0.2	4.58

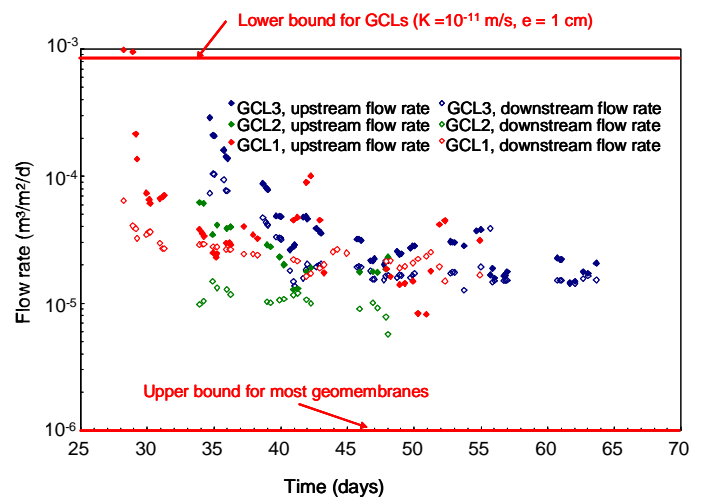


Figure 9. Synthesis of flow rates obtained for multicomponent GCLs

One can notice that the flow rate obtained through the three multicomponent GCLs is close to 10⁻⁵ m³/m²/d. This is one order of magnitude larger than the flow rate obtained for most undamaged and unaged geomembranes (10⁻⁶ m³/m²/d). It is lower than the flow rate which would be obtained for a GCL 0.01m thick with a hydraulic conductivity equal to 10⁻¹¹m/s, which is close to 10⁻³ m³/m²/d. The flow rate value in multicomponent GCLs is closer to flow rate values in geomembranes than to flow rate values in GCLs which

tends to show that the flow rate is controlled by the coating or attached film rather than by the bentonite. This result is confirmed by the fact that a change in the differential pressure between both sides of the multicomponent GCLs does not lead to significant changes in the flow rate values (Barral and Touze-Foltz 2012). Multicomponent GCLs are not behaving like porous materials confirming the trend to behave like a geomembrane. The difference in thickness between geomembranes usually tested (1mm thick as a minimum) and coatings or attached films (about 0.2 mm) can explain the difference in flow rate values observed.

Those results differ from the ones presented by Cleary & Lake (2011) for a different coated GCL. The difference arises from the density of the coating which was lower than 100g/m² for the coated GCL tested by Cleary and Lake (2011) and further tested by Barral and Touze-Foltz (2012) while the mass per unit area of the coating in the coated GCL (multicomponent GCL3) for which results are presented here is larger than 200 g/m². It is thus believed that a light coating does not allow to perfectly cover the geotextile surface resulting in “holes” in the coating that allow water to easily flow through the coated GCL. As a result in the case of light coatings the hydraulic performance is not better than the one of a GCL without coating or attached film.

2.4 Flow rates and interface transmissivity in composite liners with a GCL, including damaged coated GCLs

The work performed in the past years regarding the behaviour of geosynthetic clay liners as part of a composite liner mainly focused on the situation where the GCL is located under a hole in a high density polyethylene (HDPE) geomembrane. The flow through a defect in the geomembrane depends, as indicated by Brown et al. (1987), on the contact between the geomembrane and the underlying soil liner. According to these authors, if the contact is not perfect, once fluid has migrated through the defect, it spreads laterally through the gap existing between the geomembrane and the underlying soil, called interface. This interface flow covers an area called wetted area. Finally, the liquid migrates into and through the soil liner.

The contact between the geomembrane and the GCL was quantified in terms of flow rate through the composite liner and in terms of interface transmissivity. Various situations were tested in the past in order to evaluate the effect of a smooth geomembrane in contact with the GCL (Harpur et al. 1993, Barroso et al. 2006, 2010). Harpur et al. (1993) verified that under steady-state conditions, the most significant fraction of the flow takes place along the interface between the geomembrane and

the cover geotextile of the GCL, through the cover geotextile, and along gaps between the cover geotextile of the GCL and the bentonite. A less important amount of fluid percolates through the bentonite and below the GCL. Barroso et al. (2006, 2010) examined the influence of the hydraulic head, pre-hydration of the GCL and confining stress on the GM-GCL interface transmissivity. The results obtained by those authors showed that it is difficult to establish general trends expressing the influence of pre-hydration, confining stress and hydraulic head on the interface transmissivity. Nevertheless, it seems that, regarding the flow rate, it is important to take into account both the initial water content of the specimen and the confining stress (Barroso et al. 2006). The confining stress affects differently the flow rate, depending on the initial water content of the specimen. In fact, the flow rate in pre-hydrated GCLs is about one order of magnitude larger in tests under a confining stress of 50 kPa than in tests under 200 kPa. On the other hand, for non-pre-hydrated specimens, the flow rates are similar for the two confining stresses under steady-state flow conditions (Barroso et al. 2006).

The situation where a textured HDPE geomembrane was used in contact with the GCL was also evaluated (Barroso et al. 2008). Three different textured geomembranes were used (see Figure 10).

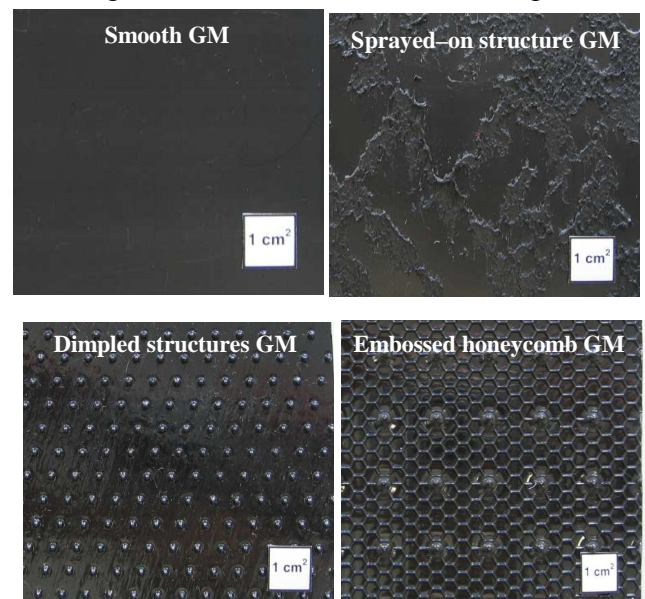


Figure 10. Types of textured geomembranes used in the study by Barroso et al. (2008)

Results showed that the tests were reproducible and that the texture had a small impact on flow rates obtained at steady-state, although, at the beginning of the tests, larger flow rates were obtained with smooth geomembranes than with textured geomembranes. This suggests that, at the early phases of the tests, the water flows more easily at the interface when smooth geomembranes are used. The texture seems to reduce the space availa-

ble at the interface for the water flow. However, with time, the sodium bentonite in the GCL swelled resulting in a better contact between the geomembrane and the GCL.

More recently, the effect of the nature of the bentonite in the GCL, sodium or calcium bentonite, leading to different flow rates in the GCL was evaluated (Mendes et al. 2010). Those authors concluded that the nature of the bentonite and the manufacturing process of the GCLs studied did not affect the GM-GCL interface transmissivity when steady-state flow conditions were reached. They did also notice that for hole diameters in the range 4 to 10 mm the diameter of the hole in the geomembrane did not significantly influence the flow rate through the GM-GCL composite liner: the expansion of the sodium bentonite was effective in blocking the puncture in the geomembrane, yielding to a significant reduction on the flow rate. The results suggest that GCLs containing sodium bentonites, whose hydraulic conductivity increases due to cation exchange, can still maintain a good performance in a composite liner in terms of GM-GCL interface transmissivity and flow rate through the composite liner.

As far as multicomponent GCLs are concerned a study was recently undertaken aiming at quantifying if the flow rates obtained through multicomponent GCLs in case the coating or attached film is damaged were in the range of values obtained for more classical composite liners with a GCL (Banour et al. 2012). Figure 11 gives a view of the four multicomponent GCLs tested. Multicomponent GCLs 1 and 3 are identical to the ones presented in Section 2.3 for the measurement of flow rate through multicomponent GCLs. Results obtained at steady-state are in the range of previous values for geomembrane-GCLs interfaces.

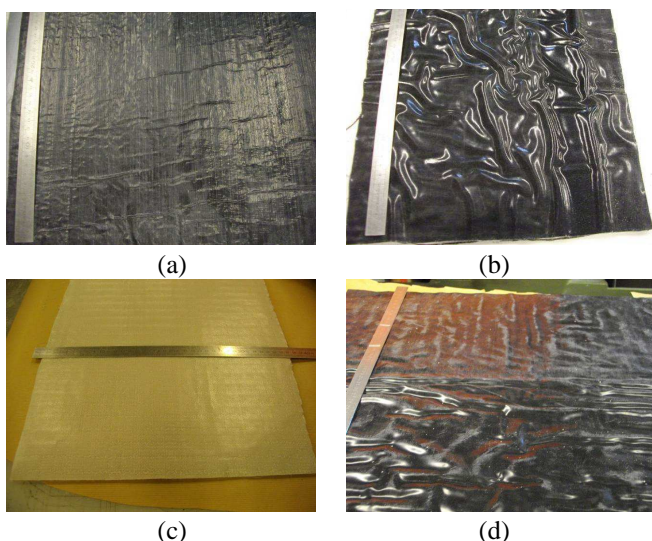


Figure 11. View of the surface of the various coated GCLs studied: (a) GCL1, (b) GCL2, (c) GCL3, and (d) GCL4.

Figure 12 gives a synthesis of the various interface transmissivity data obtained for the various

studies presented previously. All data are located under the geomembrane-GCL contact condition defined by Barroso (2005) linking the interface transmissivity to the hydraulic conductivity of the GCL in the way expressed by Equation 2:

$$\log \theta = -2.2322 + 0.7155 \log k_L \quad (2)$$

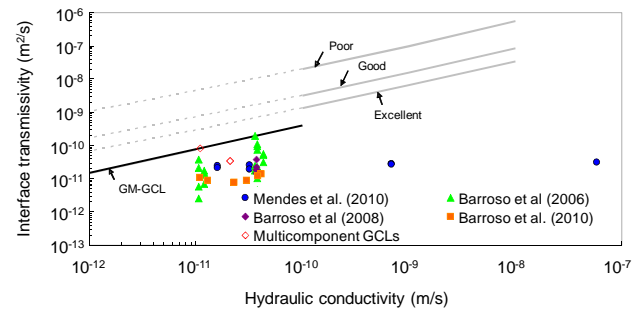


Figure 12. Synthesis of transmissivity data obtained in the literature for GCLs in contact with geomembranes and for multicomponent GCLs.

3 LEACHATE COMPOSITION FROM LANDFILLS

Besides gaseous emissions, especially methane and trace gases caused by the degradation of organic wastes, leachate emissions from landfills state the main potential long-term environmental threat (Kjeldsen et al. 2002). Traditionally, leachate analyses as a part of regular landfill monitoring have focused on nitrogen contents, oxygen consumption measured as BOD (Biological Oxygen Demand) or COD (Chemical Oxygen Demand), heavy metals and “classical” persistent organic pollutants (“POPs”) such as dioxins or polychlorinated biphenyls (PCB) (Van Praagh et al. 2011).

Metals and metalloids are still recognized as priority pollutants. In opposition to most of organic pollutants, metals and metalloids cannot be degraded in a landfill. They are thus maintained in the landfill and further mobilized in liquid or gaseous phases. The ranges of concentrations in leachate reported in the literature are considerable such as for Cd, 0.2–20 mg/L, Cr, 5–600 mg/L, Mn, 0.01–70 mg/L, Fe, 0.3–220 mg/L (Pinel-Raffaitin et al. 2006).

Landfills contain micropollutants which can have toxic effects (acute toxicity, genotoxicity, reproductive toxicity etc.) (Sisinnio et al. 2000, Takigami et al. 2002). The presence of organic contaminants in the leachate from municipal solid waste landfills has been clearly demonstrated in several countries (Oman & Hynning 1993, Ahel & Tepic 2000, Robinson et al. 2001, Hiroshi et al. 2002).

Van Praagh et al. (2011) conducted a literature review on Swedish and international literature re-

garding analytical techniques, as well as results from analysis and quantification of several single compounds or groups of emerging organic pollutants in leachate.

In 11 studies on organic substances in landfill leachate in various countries a total of 592 different compounds were identified. The major part of the identified substances can be categorized into the following groups (in descending order of number of detections): phenolic compounds, aromatic hydrocarbons, heterocyclic substances, carboxylic acids, phthalates, anilines, aliphatic acids, fenoxo

acids, organo phosphorous substances, terpenoids and triazines, some of which are used as pesticides (see Figure 13).

It is thus of the primary importance to quantify the transfer of emerging organic pollutants from leachate to the surrounding environment. This point will subsequently be discussed in Sections 4 and 5.

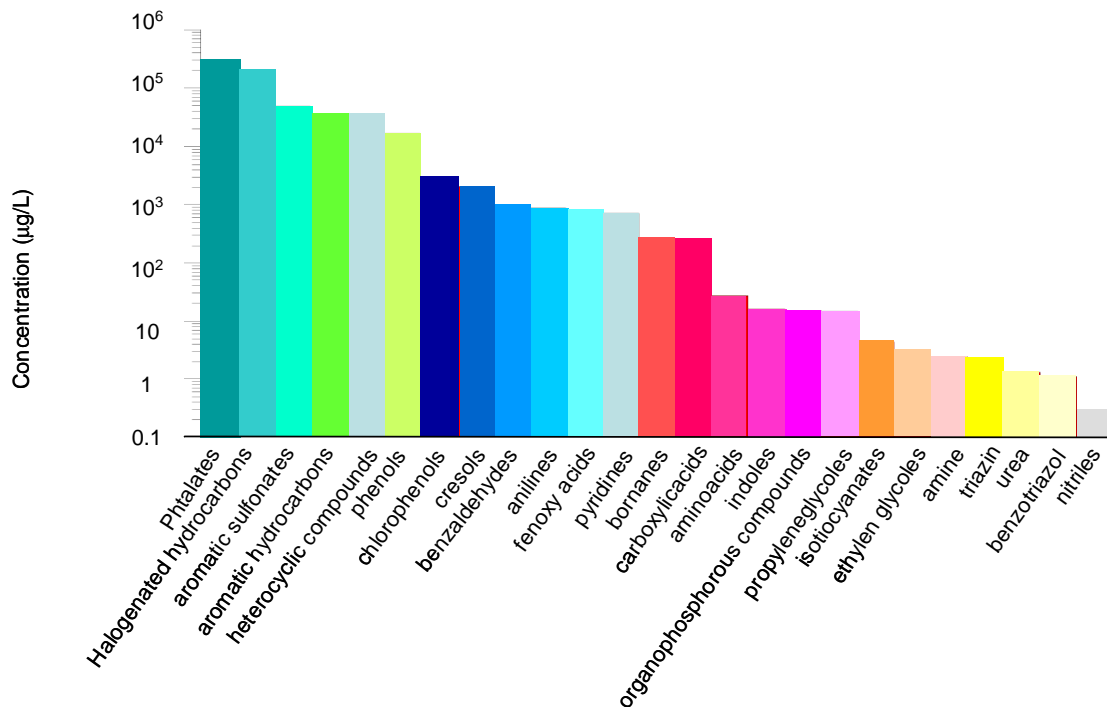


Figure 13. 20 highest detected concentrations of any compound per compound family in studies on non-Swedish landfill leachate, adapted from Van Praagh et al. (2011).

4 CHEMICAL COMPATIBILITY BETWEEN LEACHATE AND BENTONITE IN GCLS

The chemical compatibility between GCLs and leachate has been addressed in a number of studies, following the evolution with time of the hydraulic conductivity of the GCL. Syntheses of those studies can be found in Rowe (1998), Touze-Foltz et al. (2008) and Rowe (2012) for example. They will not be repeated here. The main findings can be synthesized as follows:

- When GCLs are permeated with salt solutions or simulated or real municipal solid waste (MSW) leachate, the confining stress at the time of hydration and the hydrating fluid can have a significant effect on the final hydraulic conductivity, and
- The chemical composition of the permeating fluid can have a very significant effect on the hydraulic conductivity of a GCL. In general,

the highest hydraulic conductivity and the lowest swelling were obtained in concentrated solutions or solutions with a preponderance of divalent cations. However most studies have shown that GCLs maintain low hydraulic conductivity in the range 1×10^{-12} to 1×10^{-10} m/s when permeated with real municipal solid waste (MSW) leachate, hazardous waste (HW) leachate, acid rock drainage (ARD) solutions or simulated fly ash leachate.

A question which is often raised in Europe when one has to face a variety of different bentonites in GCLs is to select the best one for a given application. Will a natural sodium bentonite perform better than a calcium activated bentonite?

Guyonnet et al. (2009) intended to study this point by permeating the 8 GCLs previously mentioned in Section 2.2.2 with a 10^{-3} M NaCl solution and then permeated with synthetic or real landfill leachate at confining stresses of 25, 50 and 100 kPa. When permeated with real leachate the values of k_L were lower than that for synthetic leachate

with $k_L \leq 4.4 \times 10^{-11}$ m/s at 100 kPa for all natural or activated sodium bentonites as shown on Figure 14. Thus no clear distinction can be made from those elements between natural sodium and calcium activated bentonites as regards the performance of GCLs towards real leachate or synthetic leachate. Values of flow rates obtained with the two calcium bentonites under testing were significantly larger.

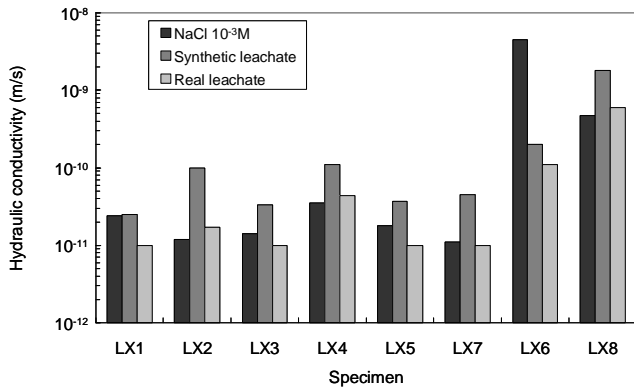


Figure 14. Values of hydraulic conductivity measured by Guyonnet et al. (2009) on 8 different GCLs under a 100 kPa load with various fluids

Those eight GCLs were analysed with a variety of indicators, not only the hydraulic conductivity, in order to help identify indicators for a better selection and control of GCLs containing sodium bentonite (including natural sodium bentonites and calcium activated bentonites).

The values of indicators were selected primarily to discard GCLs that would clearly be unsuitable for landfill bottom barrier applications, such as LX6. The value of limit free swell index is taken equal to the one which is frequently reported in data sheets thus 24 mL/2g of bentonite. A free swell index value of at least 24 mL/2g should guarantee a proportion of sodium in excess of 40–50%. The cation exchange capacity (CEC) was found to be a good indicator of the proportion of smectite and is relatively easy to measure compared to X-ray diffraction (XRD) analysis. It is expected that for bentonites in GCLs used in landfill applications in Europe, the value of a minimum CEC of 70 meq/100g should guarantee a smectite content of at least 60% in weight.

A limit value for calcite weight percent of 5% was proposed on the basis that upon total dissolution, this proportion of calcite has the potential to liberate enough calcium cations to saturate the entire cation exchange capacity of a bentonite with a CEC on the order of 75 meq/100 g. It is thus felt that the calcite weight percent is an indicator of potential difficulties that may occur in the long run.

Such performance-based indicators may contribute to pulling market quality upwards, by helping landfill operators distinguish between good

products and products that are not suitable for landfill barrier applications.

Rosin-Paumier & Touze-Foltz (2012) did further evaluate the impact of four fluids sensed as potential efficient recirculation fluids on the hydraulic conductivity of three different GCLs: two leachates originating from two areas of the same landfill, an acidogenic leachate (AL) and a methanogenic leachate (ML), a sewage sludge (SS) and leachate from a green waste repository (GL). The objective of the study was to check if the same GCL always performs best for all fluids in contact. Rosin-Paumier et al. (2010a,b) investigated the use of a filter press as a future way to quickly evaluate the potential interaction between a bentonite and a synthetic leachate (SL). The filter press test is similar to the fluid loss test (ASTM 2001) except that the effluent is collected along the test in order to acquire a filtration curve which allows calculating the permittivity of the bentonite cake formed during testing (Pantet & Monnet 2007). Rosin-Paumier et al. (2010a,b) concluded that the filter press tests may provide an indication on the fluid-bentonite interaction which can be correlated to the oedopermeameter tests results. Through the study performed thanks to filter press tests, different effects of a given fluid could be observed on the different bentonites as can be noticed on Figure 15. One can observe that for real leachate, the larger the electrical conductivity the larger the permittivity. One can also observe that different fluids give rise to different performance of the GCLs: if LX5 is the GCL behaving best with SS, AL and SL, it is not the case with ML and GL. As a consequence, the nature of the bentonite shall be adapted to the fluid to contain.

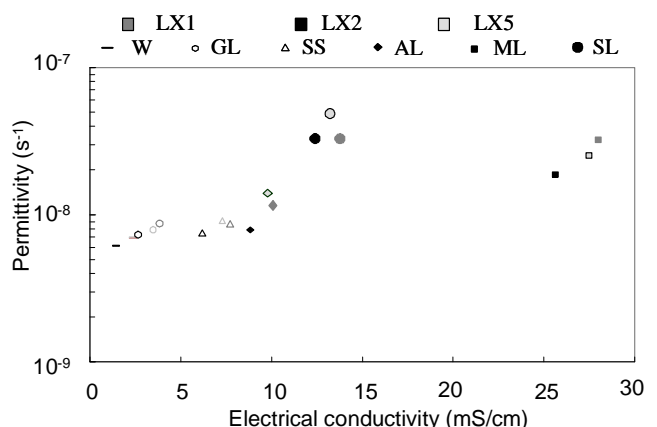


Figure 15. Permittivities obtained with the filter press under a 700 kPa air pressure according to the electrical conductivity of the dispersion. SL and W points were performed by Rosin-Paumier et al. (2010b).

ML lead to the largest permittivities of all measured with filter press tests. This result can be surprising as the calcium concentration of this leachate was lower for example than the one of the

acidogenic leachate. However, results obtained tended to show that not only the concentration in divalent cations but also the electrical conductivity of the leachate shall be taken into account to predict the effect of a leachate on a bentonite.

5 QUANTIFICATION OF DIFFUSIVE TRANSFERS THROUGH LINER MATERIALS

5.1 Theory for diffusive transfers

5.1.1 Diffusion through GCLs

Rowe & Booker (1987) developed a model for predicting one-dimensional contaminant transport through soils of finite thickness and to account for realistic landfill parameters such as surface boundary concentrations changing with time as is the case for MSW landfills. Lake & Rowe (2004), Rowe et al. (2005) and Rosin-Paumier et al. (2011) applied this model to predict one-dimensional contaminant transport for a single reactive solute without degradation through a saturated GCL, which utilizes Equation 3:

$$n \frac{\partial C}{\partial t} = nD_e \frac{\partial^2 C}{\partial z^2} - \rho_d K_d \frac{\partial C}{\partial t} \quad (3)$$

where C = concentration in the GCL at depth z and time t ; n = total porosity of the GCL; D_e = effective diffusion coefficient; ρ_d = dry density; and K_d = sorption coefficient.

Diffusion coefficients can be estimated by solving Equation 3 in combination with finite mass boundary conditions (see Rowe et al. 2004) for the test set-up. Finite mass refers to the fact that the concentration is changing throughout time at both boundary conditions, owing to mass transfer through the GCL and any sampling of the laboratory tests. K_d the sorption coefficient is first determined thanks to batch adsorption tests (Lake & Rowe 2004, Rowe et al. 2005, Ganne et al. 2008, Ahari et al. 2011).

From the knowledge of sorption coefficients on the various components of a GCL (geotextiles, geotextile fibers in the bentonite, bentonite) an equivalent sorption coefficient, K_{deq} , for the entire GCL can be calculated according to the solution suggested by Rowe et al. (2005), taking into account sorption on the geotextile, the bentonite and geotextiles fibers in the bentonite.

A global diffusion coefficient for the whole GCL can then be recalculated for each experiment and each contaminant, treating the whole GCL as a homogeneous material.

5.1.2 Theory for diffusive transfers through geomembranes

Diffusion of vapor or aqueous permeants through geomembranes occurs in three steps: adsorption, diffusion, and desorption. First, the contaminant partitions between the source medium and adjacent surface of the geomembrane. Second, the compound diffuses through the geomembrane driven by chemical potential. Finally, the compound partitions between the outer geomembrane surface and the receiving medium (Sangam & Rowe 2001).

When a geomembrane is immersed in a fluid containing a contaminant of interest for sufficient time, equilibrium is reached between the concentration in the geomembrane, c_g , and the concentration in the fluid, c_f . The two concentrations can be related by Henry's law:

$$c_g = S_{gf} c_f \quad (4)$$

where S_{gf} = partition coefficient. S_{gf} can be calculated from batch sorption tests (Sangam & Rowe 2001).

The diffusion of organic compounds through a geomembrane can be modelled by Fick's first law:

$$f = -D_g (dc_g/dz) \quad (5)$$

Where f = mass flux or permeation rate per unit area ; D_g = diffusion coefficient of organic compounds through the geomembrane; c_g = concentration of compound in the geomembrane; and z = distance parallel to the direction of diffusion. The change in contaminant concentration at any point in the geomembrane with time, t , is governed by the following differential equation according to Fick's second law:

$$\frac{\partial c_g}{\partial t} = D_g \frac{\partial^2 c_g}{\partial z^2} \quad (6)$$

The desorption is also described by Henry's law. The partition coefficient into the geomembrane is usually equal to the partition coefficient out of the geomembrane when the source and receptor fluid are the same (Sangam 2001). Since it is very difficult to measure the concentration of contaminant inside the geomembrane, consideration of mass transfer across a geomembrane in terms of the concentration in the fluids on either side of the geomembrane is used to infer the permeation characteristics of the geomembrane.

The flux associated with the diffusion process can also be written according to Equation 7:

$$f = -P_g \frac{dc_f}{dz} \quad (7)$$

Where P_g = permeation coefficient or mass transfer coefficient (m^2/s).

The diffusion coefficient can be obtained from double compartment tests (Islam & Rowe 2009, McWaters & Rowe, 2008, 2010; Sangam & Rowe 2001, Sangam & Rowe, 2005, Touze-Foltz et al. 2012) or immersion tests (Park et al. 2012).

5.2 Absorption on GCLs components

5.2.1 Case of organic pollutants

5.2.1.1 Adsorption on geotextiles

In order to investigate the possible use of geotextiles for the retention of pesticides in agricultural watersheds, Boutron et al. (2009) performed batch adsorption and desorption tests of diuron, isoproturon and azoxystrobin onto commercially available geotextiles. As far as polymeric fibers are concerned, polyamide, polyester and polypropylene were studied. Polyamide showed a high ability to sorbe diuron, with small desorption. The absorption on polypropylene and polyester was less significant but non negligible (15% of the initial mass of pesticide in water for isoproturon and 30 % for azoxystrobin).

Lake & Rowe (2004), Rowe et al. (2005) and Ganne et al. (2008) have addressed the potential for VOCs retention on geotextiles from GCLs.

The sorption isotherms are linear for VOCs and values of sorption coefficients range from 7 to 20 for 1,2-dichloroethane, 79-102 mL/g for trichloroethylene, 20 to 41 mL/g for benzene, 87 to 135 mL/g for toluene, 229-248 mL/g for ethylbenzene, 263-298 mL/g for m&p-xylene, 163-192 mL/g for o-xylene. Those values were obtained at 22°C. Rowe et al. (2005) showed that diffusion and sorption depend on temperature and both parameters were lower at 7°C than at 22°C for BTEX (benzene, toluene, ethylbenzene, m&p-xylene, and o-xylene).

Ahari et al. (2011) studied the sorption of 13 phenolic compounds, namely phenol, o-cresol, p-cresol, 2-chlorophenol (2-CP), 4-chlorophenol (4-CP), 2,4-dimethylphenol (2,4-DMP), 3,4-dimethylphenol (3,4-DMP), 2,4-dichlorophenol (2,4-DCP), 2,4,6-trichlorophenol (2,4,6-TCP), 2,3,5,6-tetrachlorophenol (2,3,5,6-TeCP), 2,3,4,6-tetrachlorophenol (2,3,4,6-TeCP), pentachlorophenol (PCP), and bisphenol A (BPA).

They noticed that the adsorption isotherms are non-linear contrarily to what was observed in the case of VOCs (Rowe et al. 2005, Ganne et al. 2008). The Freundlich model was thus used in this study to model adsorption. However for the sake of comparison to previous values obtained with VOCs Ahari et al. (2011) calculated also K_d values under the hypothesis that sorption was linear. All obtained values of K_d ranged between 2.7 and 8.9 mL/g for compounds studied. These values remain

low compared to values obtained with VOCs. An important observation is that the amount of chlorophenols sorbed on geotextiles increases with an increasing number of chlorine atoms.

5.2.1.2 Adsorption on bentonite

Lake & Rowe (2004) have addressed the potential for volatile organic compounds retention on bentonites from GCLs. Very low values of partition coefficients were obtained in the literature (below 1 mL/g).

A few studies have recently addressed the potential for sorption of some phenolic compounds on bentonites or organobentonites (Banat et al. 2000, Yoo et al. 2004, Hameed 2007, Richards & Bouazza 2007, Malusis et al. 2010, Ahari et al. 2011). In the study by Ahari et al. (2011) adsorption curves obtained with bentonite are non linear. The Freundlich model was thus also used for bentonite. Results obtained were consistent with previous results from Banat et al. (2000) for phenol. In order to compare with other data from the literature where linear adsorption isotherms were obtained, an adsorption coefficient, K_d , was also determined in the case of the adsorption of phenol on bentonite under the hypothesis that the isotherm was linear. Values obtained following the assumption of a linear isotherm ranged between 2.5 and 2.6 mL/g and felt well within the range given by Richards & Bouazza (2007) ($K_d = 1-5$ mL/g) and by Haijian et al. (2009) ($K_d = 1.2-3.3$ mL/g) for phenol. According to what was observed for geotextiles, the adsorption of chlorophenols increased with an increasing number of chlorine atoms. This study also showed that contrarily to what was observed for VOCs, the difference in the adsorbed amount of phenolic compounds between the geotextile and bentonite is less pronounced.

5.2.2 Case of inorganic pollutants

Lange et al. (2004) examined the migration of various metals (Al, Fe, Mn, Ni, Pb, Cd, Cu, Zn) through GCLs in the presence of a synthetic municipal solid waste leachate. GCLs were found to be capable of retarding metals, however at specific pH conditions. Mn behaved similarly to Cl, indicating that it was the least attenuated metal. Al, Fe and Cu were highly retarded and retained within the clay. Ni, Zn, and Cd were moderately attenuated. Ca could have also been responsible for the lack of metal retention of the leachate species. Due to the higher retention at higher pHs and the release of metals at lower pHs, adsorption of hydrolyzed species in addition to cation exchange were hypothesized as the mechanisms most responsible for metal retention.

5.3 Diffusive transfers through GCLs

5.3.1 Diffusion of inorganic species through GCLs

Lake & Rowe (2000) have shown a linear increase of the diffusion coefficient of sodium and chloride with an increase in the bulk GCL void ratio, for void ratios in the range 1.5 to 3. The bulk GCL void ratio was first defined by Petrov et al. (1997) as:

$$e_b = \frac{H_{GCL} - H_s}{H_s} \quad (8)$$

Where H_{GCL} = GCL height; and H_s = height of solids in the GCL. H_s is defined by Equation 9:

$$H_s = \frac{M_{bent}}{\rho_s(1 + w_0)} + \frac{M_{geo}}{\rho_{sg}} \quad (9)$$

Where M_{bent} = mass of bentonite per unit area in the GCL; M_{geo} = mass of geosynthetics per unit area in the GCL; ρ_s = density of bentonite solids, ρ_{sg} = density of polypropylene geotextile solids; and w_0 = initial water content of the bentonite.

The diffusion coefficients of sodium and chloride deduced from GCL diffusion tests performed with 3 g/L to 5 g/L decreased linearly with decreasing final bulk GCL void ratio. The diffusion coefficient was shown to be dependent on the source solution and, when the NaCl concentration was increased significantly, the diffusion coefficient deduced was also shown to increase. The diffusion coefficients were estimated to be in the range 10^{-10} to 2×10^{-10} m²/s.

Lange et al. (2009) studied the diffusion of various metals for the following four cases where a GCL might serve as an effective barrier material to metals and metalloids: acidic rock drainage, gold mine tailings, lime-treated mine effluent, and municipal solid waste. The average diffusion coefficients for Cu, Cd, Zn, Fe and Ni covered a narrow range from 0.67×10^{-10} to 0.89×10^{-10} m²/s. The diffusion coefficients for As, Al, Mg, Mn and Sr were in the range 0.80×10^{-10} to 1.6×10^{-10} m²/s. The individual metal diffusion coefficients did not change significantly for different solution compositions. This suggested that although solution composition had some effect on the metal diffusion coefficient, sorption to the GCL was the dominant control on metal mobility.

5.3.2 Diffusion of organic species through GCLs

The quantification of diffusion mechanisms of VOCs) was undertaken for virgin GCLs (Lake & Rowe 2004, Rowe et al. 2005, Ganne et al. 2008) containing sodium bentonite.

Lake & Rowe (2004) did not observe a significant increase in the diffusion coefficient for bulk GCL void ratios in the range 4.1 to 4.8. They indicated that bulk GCL void ratios in the range 4.1 to 4.8 corresponded to low normal stresses and that the corresponding diffusion coefficients represented an upper bound of diffusion coefficient values.

Rosin-Paumier et al. (2011) did further study the effect of the nature of the bentonite (natural sodium versus calcium activated) on the diffusion of VOCs and did not notice any significant impact of the nature of the bentonite.

The diffusion coefficients for VOCs in bentonite range between 1 and 3×10^{-10} m²/s at 23°C for bulk GCL void ratios in the range 3.7 to 4.8.

5.3.3 GCLs after cation exchange

Rosin-Paumier et al. (2011) did submit a GCL containing natural sodium bentonite to cation exchange by permeating a synthetic leachate containing a mixture of monovalent and divalent cations in proportions representative of what a real landfill leachate would contain. Cation exchange did result in an increase in the hydraulic conductivity of the GCL specimen by a factor 8.5.

Diffusion was then performed on three specimens cut in the sample submitted to cation exchange. An increase in the diffusion coefficient was observed as compared to the virgin GCL specimens for a bulk GCL void ratio equal to 3.9.

The largest increase in diffusion coefficient was observed for TCE as compared to the other contaminants with an increase in the diffusion coefficient obtained equal to 2.6 (from 1.0×10^{-10} to 2.6×10^{-10}). For DCM and DCA the increase was of a factor 1.4 respectively from 2.3×10^{-10} to 3.1×10^{-10} m²/s and from 1.9×10^{-10} to 2.6×10^{-10} m²/s. This ratio is not as large as the increase in the hydraulic conductivity of the GCL.

It was further observed that a decrease in the bulk GCL void ratio from 3.9 to 3 would annihilate the effect of cation exchange so that the diffusion coefficient would not increase for DCM and DCA. Such a decrease could be obtained along the life of the landfill as the height of waste increases. A detrimental effect of cation exchange on the hydraulic conductivity of a GCL is thus not necessarily indicative of a detrimental effect on the diffusion coefficient of VOCs through GCLs. This result has to be confirmed for other GCLs and other pollutants.

5.4 Diffusive transfers through geomembranes

5.4.1 Diffusion of inorganic species

Rowe (2005) presented the results of a diffusion test of chloride that had at the time of publication run for 12 years. The receptor concentration in this tests remained below about 0.02% of the source

concentration, lying within the range of analytical uncertainty for the chemical analysis. Rowe (2005) also mentions a study by August & Tatzky concluding to negligible diffusion of heavy metals salts from a 0.5M acid solution through HDPE over a 4 year test period.

From those results it is concluded that an intact GM is an excellent barrier to the advective and diffusive migration of inorganic contaminants from a leachate (Rowe 2012).

5.4.2 Diffusion of organic species

August & Tatzky (1984) found that strongly polar penetrant molecules have permeation rates in a HDPE geomembrane in the range 7×10^{-7} (for methanol) to 9.4×10^{-6} $\text{m}^3/\text{m}^2/\text{d}$ for trichloroethylene.

A number of studies regarding the use of geomembranes focused on the diffusion of volatile organic compounds (VOCs) for virgin HDPE geomembranes (Park & Nibras 1993, Prasad et al. 1994, Müller et al. 1998, Sangam & Rowe 2001, Touze-Foltz et al. 2011, Park et al. 2012) virgin PVC, LLDPE with and without a co-extruded ethylene vinyl-alcohol (EVOH) inner core geomembranes (McWaters & Rowe 2008, 2010), fluorinated HDPE geomembranes (Sangam & Rowe 2005) and aged HDPE geomembranes (Rowe et al. 2003, Islam & Rowe 2009).

Diffusion coefficients of VOCs in virgin HDPE geomembranes range from 0.37 to 22.8×10^{-13} m^2/s for benzene and dichloromethane respectively with partition coefficients ranging from 1.8 to 189. The resulting permeation coefficient lies between 1 and 70×10^{-12} m^2/s .

Sangam & Rowe (2005) examined the effect of surface fluorination of an HDPE geomembrane on diffusion of VOCs. The surface fluorination consisted of applying elemental fluorine, which exchanged with hydrogen along polymer chains at the surface of a polyolefin substrate. The partition coefficient was shown to remain essentially the same after the surface fluorination; however, the surface fluorination resulted in a reduction in both the diffusion and the permeation coefficients by factors ranging between 1.5 and 4.5, depending on the hydrocarbon examined.

McWaters & Rowe (2010) studied the diffusive migration in the aqueous and vapour phases benzene, toluene, ethylbenzene and xylenes through a 0.76 mm PVC-P geomembrane and 0.76 mm thick linear low-density polyethylene (LLDPE). Diffusion coefficients for the PVC geomembrane were in the range 5 to 10×10^{-13} m^2/s for diffusion from both the aqueous and vapour states. The range for LLDPE was 2.5 to 5×10^{-13} m^2/s . The partition coefficients for PVC were in the range 100–1075 with respect to aqueous phase concentrations. The corresponding values with respect to vapour phase

concentrations were 22–290. LLDPE partition coefficients were 200–475 with respect to aqueous phase concentrations and 44–123 with respect to vapour phase concentrations. The resulting permeation coefficients are thus in the range $130\text{--}750 \times 10^{-12} \text{m}^2/\text{s}$ for PVC and $60\text{--}110 \times 10^{-12} \text{m}^2/\text{s}$ for LLDPE.

McWaters & Rowe (2010) did further study the diffusive properties of two coextruded geomembranes, one with a polyamide inner core and the other with an ethylene vinyl-alcohol (EVOH) inner core, and a standard 0.53-mm LLDPE geomembrane. Results indicate a significant reduction in mass flux through the coextruded geomembranes compared to conventional LLDPE. The EVOH coextruded geomembrane had the lowest permeation coefficients D about 8×10^{-15} m^2/s for diffusion from the aqueous phase. These values for EVOH are upper bounds and the actual values may be even lower. The polyamide coextruded geomembrane had higher values than for EVOH, with a D range of 5 to 8×10^{-14} m^2/s from the aqueous phase. The highest diffusion coefficients were for the standard LLDPE, which ranged from 2 to 4×10^{-13} $\text{m}^2 \text{s}^{-1}$. The partition coefficients for EVOH were in the range 160–700 with respect to aqueous phase concentrations. The polyamide coextruded geomembrane partition coefficients were 120–430 and the LLDPE partition coefficients were 180–450 with respect to aqueous phase concentrations. The resulting permeation coefficients are thus in the range $2\text{--}6 \times 10^{-12} \text{m}^2/\text{s}$ for EVOH, $7\text{--}22 \times 10^{-12} \text{m}^2/\text{s}$ for the polyamide coextruded geomembrane and $60\text{--}200 \times 10^{-12} \text{m}^2/\text{s}$ for LLDPE.

The EVOH geomembrane did thus show a 5–12-fold decrease in the permeation coefficient in comparison to a 2.0-mm high density polyethylene geomembrane.

Theoretical laws to predict the evolution of the diffusion coefficient, the partition coefficient and the permeation coefficient with the solubility, octanol-water partition coefficient and molecular diameter of various VOCs were given in the literature for the diffusion of VOCs through virgin HDPE geomembranes (Sangam & Rowe 2001, Joo et al. 2004, Park et al. 2012).

The diffusion of Polymbriomined Diphenyl Ethers (PBDE) (Saheli et al. 2011) is currently under study.

Touze-Foltz et al. (2012) studied the diffusion of phenol, *o*-cresol, *p*-cresol, 2,4-xylenol, 3,4-xylenol, 2-chlorophenol, 4-chlorophenol, 2,4-dichlorophenol, 2,4,6-trichlorophenol, 2,3,4,6-tetrachlorophenol, 2,3,5,6-tetrachlorophenol, pentachlorophenol and bisphenol A (BPA) through a virgin HDPE geomembrane.

The results show that the partition coefficient is linked to the aqueous solubility and the *n*-octanol/water partition coefficient of the contami-

nant. This latest result is logical as the less polar the contaminant is, the less it is soluble in water. No clear link could be evidenced with the molecular diameter when all phenolic compounds were taken into account (especially BPA). However a good correlation was observed for chlorophenols between molecular weight and partition coefficient which may be related to polarity.

An analysis of the sole chlorophenols (2-CP, 4-CP, 2,4-DCP, 2,3,6-TCP, 2,3,4,6-TeCP, 2,3,5,6-TeCP et PCP) tends to show also that the partition coefficient is closely linked with the degree of substitution of chlorine atoms on the phenolic nucleus, phenomenon which may also be attributed to a difference in polarity of the various chlorophenols studied.

The partition coefficient values of phenolic compounds obtained were low compared to those of VOCs whose chemical structure is similar (benzene compared to phenol and toluene compared to o-cresol and p-cresol), tending to show that the partition coefficient decreases as the hydroxyl group tends to make the molecule more soluble.

As regards the diffusion coefficient a good correlation with the aqueous solubility and the n-octanol/water partition coefficient of the phenolic compounds is observed as shown on Figure 16. A good correlation with the molecular diameter is also obtained when one disregards the values obtained for BPA.

Those trends are consistent with previous trends obtained for VOCs. However, the range of parameters obtained is significantly different so that the empirical equations derived for VOCs and available in the literature are not valid to predict the evolution of the permeation coefficient (see Figure 17) of phenolic compounds except as regards the evolution with the molecular diameter when disregarding the parameters obtained for BPA. Those trends require further confirmation for other chemical families before one can undertake the prediction of the diffusion coefficient based on the sole value of the molecular diameter for molecular diameter close to 0.5nm.

5.4.3 *Effect of geomembrane ageing on diffusion*

The effects of aging of HDPE geomembranes on the diffusion and partition of benzene, toluene, ethylbenzene, and xylenes were examined by Islam & Rowe (2009). Two different 1.5 mm thick HDPE geomembranes were aged in the laboratory at 85°C by immersing in a synthetic leachate for up to 32 months. Partition and diffusion tests were performed at room temperature on both unaged and aged geomembranes using a dilute aqueous solution. The diffusion and partition coefficients decreased with increased aging.

The calculated permeation coefficients decreased by 36–62% after aging the geomembrane

for about 10–32 months. This decrease in diffusion, partition, and permeation coefficients is related to the increase in geomembrane crystallinity during aging.

Those results are consistent with previous results from Rowe et al. (2003) who showed that for a 14 years old HDPE geomembrane sampled in a leachate lagoon the permeation coefficients were between four and five times lower than values obtained for unaged HDPE geomembranes typical of present day production (no virgin specimen available).

6 CONCLUSION

The objective of this Keynote Lecture was to give an insight in the state of the art regarding measurement techniques for the evaluation of transfer properties in lining materials for landfills. An insight was first given regarding measurement techniques to quantify the advective flow rates through geomembranes and multicomponent GCLs. It was shown that the flow rate through multicomponent GCLs is larger than the flow rates for geomembranes, but significantly smaller than the flow rate for usual GCLs, as long as the coating is continuous. The effect of some parameters on the flow rate through GCLs like the nature of the bentonite, the mass per unit area, the hydration with or without load were discussed thanks to the results of recent studies. Finally, as far as advective transfers are concerned, the quantification of flow rates in composite liners or in multicomponent GCLs when the geomembrane of the film or coating is damaged was discussed. All results obtained at steady-state are in the same range, located below a geomembrane GCL contact condition previously defined in the literature. No significant effect of the texture of the geomembrane or of the nature of the bentonite in the GCL was evidenced. A brief insight was then given in the composition of leachate, especially as regards their content in microorganic pollutants. This justifies the study of the diffusion of microorganic pollutants further discussed in Section 5.

The question of the chemical compatibility between bentonite and leachate was first addressed.

Recommendations given in the literature in terms of indicators to ensure the performance of the bentonite in the GCL on the long term were given.

The need to adapt the nature of the bentonite to the leachate to contain was also discussed.

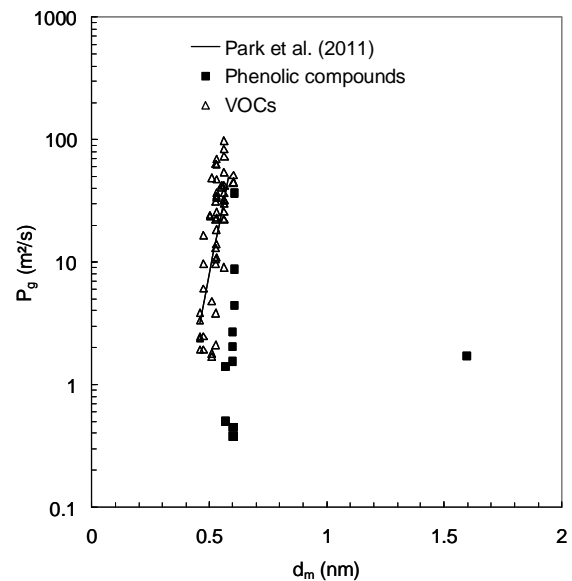
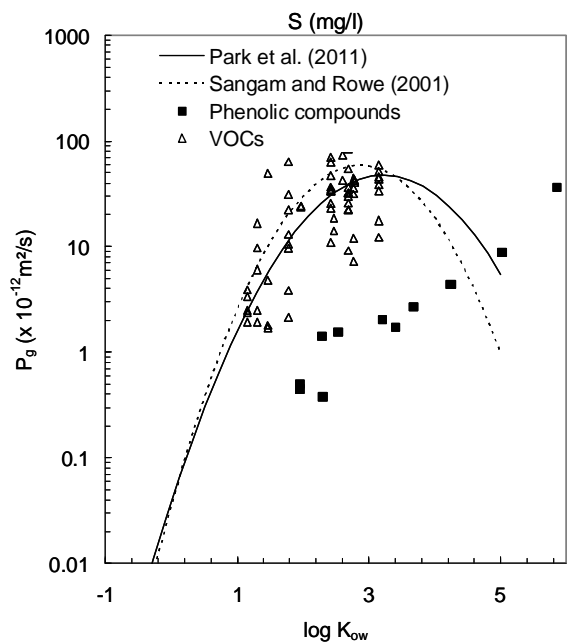
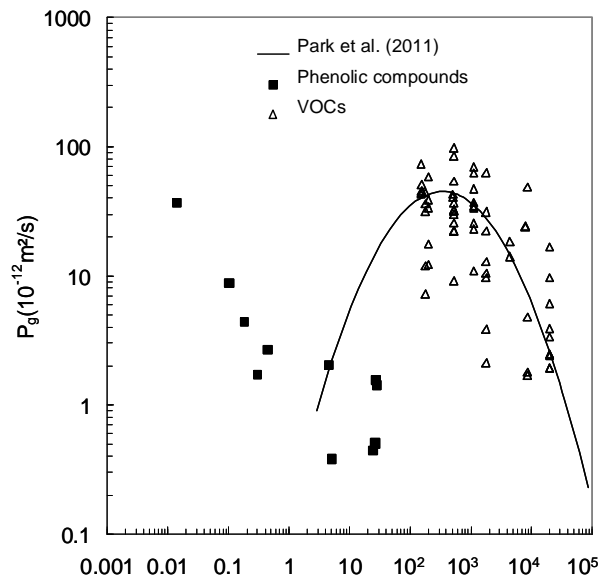
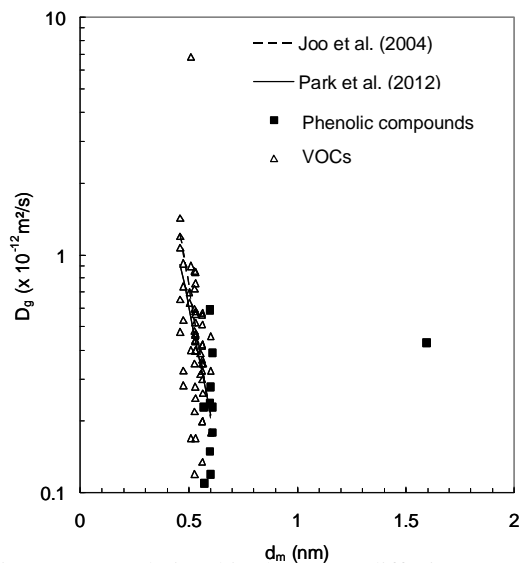
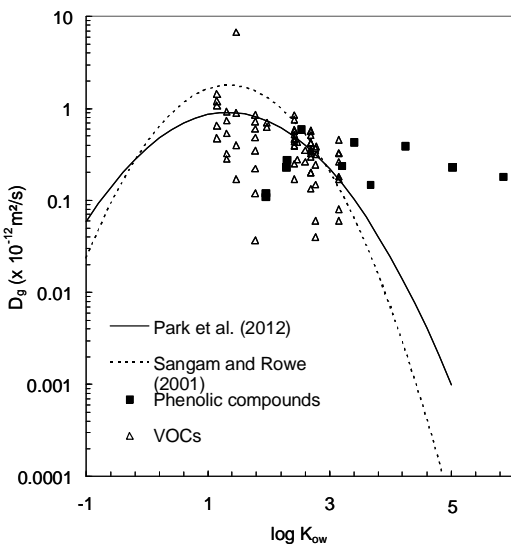
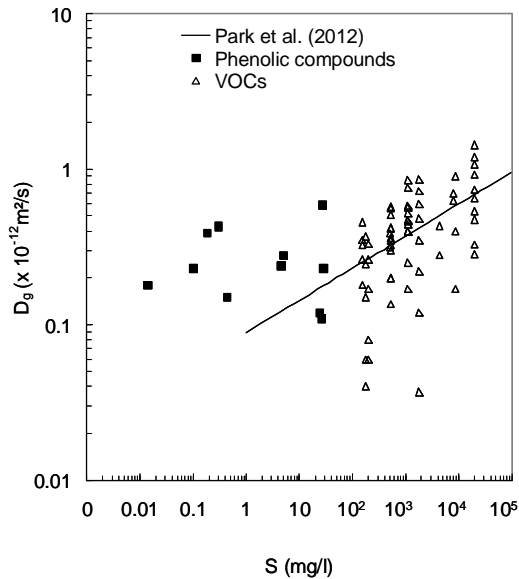


Figure 16. Relationship between diffusion coefficient, D_g , and: (a) aqueous solubility, (b) octanol-water partition coefficient, K_{ow} , and (c) molecular diameter, d_m for phenolic compounds and VOCs (adapted from Touze-Foltz et al. 2012).

Figure 17. Relationship between permeation coefficient, P_g , and: (a) aqueous solubility, (b) octanol-water partition coefficient, K_{ow} , and (c) molecular diameter, d_m for phenolic compounds and VOCs (adapted from Touze-Foltz et al. 2012).

Finally, diffusion of organic and inorganic species through GCLs and geomembranes was discussed. Very few species have been investigated until now. Data exist for various kinds of geomembranes especially as regards the diffusion of VOCs but a recent study has shown that the empirical laws used for the prediction of the diffusion of VOCs in geomembranes cannot be extended to other chemical families like the one of phenolic compounds. This emphasizes the need for further research in this field.

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