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Steady-State Analysis of Pollutant Transport To Assess Landfill Liner Performance

by

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#### **ABSTRACT**

New analytical solutions are presented for the assessment of the impact of contaminant transport through landfill liners on groundwater quality under steady-state conditions. These solutions can be applied to evaluate the equivalency and the effectiveness of landfill liners, even those that include a geomembrane, taking into account the presence of a natural attenuation layer interposed between the engineered barrier and the underlying aquifer. The impact of a contaminant on groundwater quality is quantified through the determination of the contaminant concentration along the horizontal direction of the groundwater flow for the case of thin aquifers that are only a few meters thick. For the case of thick aquifers, both a closed-form analytical solution and a step-by-step numerical solution are provided for the calculation of the variations in the contaminant concentration in horizontal and vertical directions within the aquifer.

**Keywords chosen from ICE Publishing list**: Groundwater, Numerical methods, Waste containment & disposal system

#### List of notation

 $\alpha_T$  is the transverse dispersivity within the aquifer

 $\gamma$ ,  $\Gamma$ ,  $\eta$ ,  $\eta_D$ ,  $\eta'$ ,  $\eta'_D$ ,  $\chi$  are dimensionless parameters of the analytical solutions

 $\Delta h$  is the hydraulic head difference across the landfill bottom barrier

 $\Delta X$  is the interval of the horizontal distance for the numerical solution

 $\Delta Y'$  is the interval of the vertical distance for the numerical solution

 $\theta$  is the volumetric water content

 $\Theta$  is the transmissivity of the interface between the geomembrane and the

underlying soil

D<sub>g</sub> is the diffusion coefficient of the geomembrane

D<sub>h</sub> is the hydrodynamic dispersion/diffusion coefficient of the landfill bottom

barrier

D<sub>h,x</sub> is the hydrodynamic dispersion/diffusion coefficient in the horizontal direction

within the aquifer

D<sub>h,y</sub> is the hydrodynamic dispersion/diffusion coefficient in the vertical direction

within the aquifer

D<sub>hi</sub> is the hydrodynamic dispersion/diffusion coefficient of the i-th layer of the

landfill bottom barrier

J<sub>s</sub> is the pollutant vertical mass flux coming from the landfill

 $K_g$  is the partition coefficient between the geomembrane and the solute

L is the thickness of the landfill bottom barrier

L<sub>g</sub> is the geomembrane thickness

L<sub>i</sub> is the thickness of the i-th layer of the barrier

L<sub>w</sub> is the length of the geomembrane wrinkle

N<sub>1</sub> is the number of mineral layers in the landfill bottom barrier

 $N_X$  is the number of nodes in the horizontal direction for the numerical solution

 $N_{Y'}$  is the number of nodes in the vertical direction for the numerical solution

P<sub>L</sub> is the Peclet number of the landfill bottom barrier

Q is the leakage rate through a single hole of the geomembrane

RC is the dimensionless relative concentration of the pollutant

X is the dimensionless horizontal distance below the landfill

Y, Y' are dimensionless vertical distances from the top of the aquifer

Yaq is the dimensionless depth of the aquifer

a, d, G are the dimensionless parameters of the numerical solution

b is the half-width of the geomembrane wrinkle

c is the pollutant concentration in a thick aquifer

c<sub>0</sub> is the pollutant concentration in the landfill leachate

c<sub>b</sub> is the pollutant concentration at the bottom of the landfill barrier

 $c_x$  is the pollutant concentration in a thin aquifer

 $c_{x0}$  is the pollutant concentration in the aquifer upstream from the landfill

h is the thickness of the aquifer

h<sub>b</sub> is the height of the water level at the bottom of the landfill barrier

h<sub>p</sub> is the height of the ponded leachate above the landfill bottom barrier

k<sub>eq</sub> is the equivalent hydraulic conductivity of the landfill bottom barrier

k<sub>i</sub> is the hydraulic conductivity of the i-th layer of the landfill bottom barrier

is the horizontal length of the landfill in the direction of the groundwater flow

 $n_{aq}$  is the porosity of the aquifer

n<sub>h</sub> is the number of the geomembrane defects per unit area

n<sub>i</sub> is the porosity of the i-th layer of the landfill bottom barrier

q is the vertical infiltration flux coming from the landfill

q<sub>x</sub> is the groundwater horizontal flux

 $q_{x0}$  is the groundwater horizontal flux upstream the landfill

q<sub>y</sub> is the groundwater vertical flux

r is the number of the series terms of the analytical solution for aquifers with a

finite thickness

is the time

 $v_{x0}$  is the horizontal seepage velocity of groundwater upstream from the landfill

x is the horizontal distance below the landfill

y is the vertical distance from the top of the aquifer

#### 1. INTRODUCTION

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2 The design of landfill liners is aimed at minimizing both leachate flow and contaminant transport due to advection and molecular diffusion from the waste to the groundwater system (Rowe et al., 2004; 3 Shackelford, 2014). Modern liners comprise compacted clay layers (CCLs) and/or geosynthetic 4 components, such as geomembrane layers (GMLs) and geosynthetic clay layers (GCLs). GMLs can 5 be coupled with CCLs and GCLs to form composite liners that provide optimal groundwater 6 protection. Moreover, the aquifer located beneath the landfill is typically separated from the waste 7 8 not only by these engineered barriers, but also by a natural foundation or attenuation layer (AL), 9 which plays an important role in limiting contaminant migration (Rowe and Brachman, 2004). 10 The preliminary design of a lining system generally requires the assessment of both its *equivalency* 11 with the lining system that is prescribed by the environmental regulations in force and its effectiveness with respect to specified performance criteria (Katsumi et al., 2001; Foose, 2010). One common 12 performance criterion is that the design should ensure an acceptable risk for human health and the 13 environment, which is typically quantified through a prescribed maximum value of the contaminant 14 concentration in the groundwater. For this reason, the assessment of liner performance generally 15 requires one to ascertain that the concentration of pollutants that are released by the waste remains 16 below a prescribed threshold level at a specified compliance point, which is commonly represented 17 18 by a monitoring well located in the underlying aquifer and downstream from the landfill (Figure 1). Numerical solutions implemented in software products are currently available to evaluate the 19 groundwater contaminant concentration below the landfill (Rowe and Booker, 1985a, 1985b; El-Zein 20 21 and Rowe, 2008; El-Zein et al., 2012; Xie et al. 2016). However, such numerical solutions do not exclude the interest in implementing analytical solutions, which may be considered useful analysis 22 tools, due to their simplicity and repeatability. In this paper, analytical solutions are developed under 23 the restrictive assumptions of steady-state conditions and constant source concentration in the waste 24 leachate. These conditions, which were also assumed by Guyonnet et al. (2001) and Foose (2010), 25 26 exclude the possibility of modeling time-varying properties and time-dependent phenomena, and

typically result in conservative predictions of the groundwater contaminant concentration (Shackelford, 1990; Rabideau and Khandelwal, 1998; Rowe et al., 2004). As a result, such solutions should not be considered as long-term, realistic simulations of contaminant migration, but rather as conservative estimates of the risk related to a given contaminant concentration in the waste leachate, in a similar way to a Tier-2 analysis of the ASTM risk-based corrective action (RBCA) standard (ASTM, 2015) for a polluted site. The scenarios considered in this paper include vertical pollutant transport through an engineered barrier, which can overlie a foundation or attenuation layer, and horizontal transport in the underlying aquifer (Figure 1). With respect to the already available steady-state analysis approaches (Guyonnet et al., 2001; Foose, 2010), the solutions that are derived in this paper allow the contaminant concentration distribution to be evaluated within the aquifer beneath the landfill. A first analytical solution is derived for the case of aquifers that are sufficiently thin in order to neglect the vertical distribution of the contaminant concentration in the groundwater. When the aquifers are thick and the vertical dispersion of the contaminants cannot be neglected, a two-dimensional mass balance in the aquifer has to be considered, and a set of analytical and step-by-step numerical solutions is developed. An example of the application of the proposed analytical solutions is presented to assess the migration of an organic pollutant (toluene, C<sub>6</sub>H<sub>5</sub>-CH<sub>3</sub>) for the case of composite liners, in which the mineral layers (CCL or GCL) are coupled with a GML.

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### 2. POLLUTANT CONCENTRATION IN THIN AQUIFERS

If the thickness of the aquifer, h, is no more than a few meters, the vertical component of the groundwater volumetric flux can be neglected with respect to the horizontal one. The water balance inside an aquifer volume of infinitesimal length, dx, can be expressed as follows:

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$$50 h \cdot \frac{dq_x}{dx} = q (1)$$

51

where  $q_x$  is the horizontal groundwater flux, q is the vertical (infiltration) flux coming from the landfill and x is the horizontal distance below the landfill taken in the direction of the groundwater flow (Figure 2). After integration of Eq. 1, the groundwater flux  $q_x$  varies linearly beneath the landfill as

55 56 follows:

$$q_{x} = q_{x0} + \frac{q}{h} \cdot x \tag{2}$$

58

- where  $q_{x0}$  is the groundwater flux just upstream from the landfill, i.e. at location x = 0.
- For thin aquifers, the pollutant concentration can be assumed to be invariant with the vertical position,
- and transport by advection is dominant relative to transport by longitudinal hydrodynamic
- dispersion/diffusion in the horizontal direction (Rowe and Booker, 1985a). Under steady-state
- conditions, the pollutant mass balance inside the aquifer can be obtained by combining the horizontal
- advective mass flux with the vertical mass flux derived from the landfill,  $J_s$ , as follows:

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$$66 h \cdot \frac{d}{dx} (q_x \cdot c_x) = J_s (3)$$

67

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where  $c_x$  is the pollutant concentration in the aquifer beneath the landfill (Figure 2).

69 The vertical flux of a miscible pollutant though a multi-layer barrier is given by (Manassero et al.,

70 2000; Guyonnet et al., 2001):

71

72 
$$J_s = q \frac{c_0 \cdot \exp(P_L) - c_b}{\exp(P_L) - 1}$$
 (4)

73

74 where  $c_0$  is the pollutant concentration in the leachate on the top of the barrier,  $c_b$  is the pollutant

concentration at the bottom of the barrier, which is supposed to coincide with the top of the aquifer

located beneath the landfill, and P<sub>L</sub> is the dimensionless Peclet number of the barrier.

77 The Peclet number, which represents the ratio of advective transport rate to diffusive-dispersive

78 transport, can be expressed as follows:

79

$$80 P_{L} = \frac{q}{\Lambda} (5)$$

81

where  $\Lambda$  is the equivalent diffusivity of the multi-layer barrier. For a volatile organic compound

83 (VOC), which can diffuse through GMLs, Λ is given by (Manassero et al., 2000; Guyonnet et al.,

84 2001; Katsumi et al., 2001; Foose, 2010; Shackelford, 2014):

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86 
$$\Lambda = \frac{1}{\frac{L_g}{K_g \cdot D_g} + \int_0^L \frac{dz}{\theta \cdot D_h}}$$
 (6)

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where  $L_g$  is the thickness of the geomembrane that is placed at the top of the barrier,  $K_g$  is the partition

coefficient between the geomembrane and solute, Dg is the diffusion coefficient of the geomembrane,

θ is the volumetric water content, D<sub>h</sub> is the hydrodynamic dispersion/diffusion coefficient in the

vertical direction and L is the total thickness of the barrier system. If pollutant transport occurs under

saturated conditions, the volumetric water content is equal to the porosity of the layers which constitute the barrier system, and the equivalent diffusivity can be expressed as follows:

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95 
$$\Lambda = \frac{1}{\frac{L_g}{K_g \cdot D_g} + \sum_{i=1}^{N_1} \frac{L_i}{n_i \cdot D_{bi}}}$$
 (7)

96

where L<sub>i</sub> is the thickness of the i-th layer, n<sub>i</sub> is the porosity of the i-th layer, D<sub>hi</sub> is the hydrodynamic 97 98 dispersion/diffusion coefficient of the i-th layer and N<sub>1</sub> is the number of mineral layers in the barrier 99 system (Figure 3). 100 In the absence of the GML or in the case of degradation of the GML,  $\Lambda$  can be calculated by omitting 101 the first term related to the GML at the denominator of Eq. 6 or 7. For inorganic solutes, whose 102 diffusion through GMLs is extremely slow and can be neglected with respect to the transport through GML holes, the mass flux through composite barriers, J<sub>s</sub>(Composite), can be calculated by multiplying 103 the mass flux without the GML, J<sub>s(Mineral)</sub>, by the ratio between the leakage area and the total area of 104 the barrier, expressed as the ratio between the leakage rate through the composite barrier, q<sub>(Composite)</sub>, 105 and the water volumetric flux through the mineral barrier without the GML,  $q_{(Mineral)}$ , i.e.  $J_{s(Composite)} =$ 106

In a thin aquifer, the pollutant concentration at the top of the aquifer,  $c_b$ , can be assumed to coincide with the concentration in the aquifer,  $c_x$ , as the change in pollutant concentration along the vertical direction is negligible.

 $J_{s(Mineral)} \cdot q_{(Composite)}/q_{(Mineral)}$ , as proposed by Katsumi et al. (2001) and Foose (2010).

As a result, when Eq. 4 is inserted into Eq. 3, and the variation of  $q_x$  along the x-direction, which is given by Eq. 2, is taken into account, the pollutant mass balance yields the following first-order differential equation:

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$$\frac{dc_x}{dx} + \chi \cdot \left(\frac{q}{q_{x0} \cdot h + q \cdot x}\right) \cdot c_x = \chi \cdot \left(\frac{q}{q_{x0} \cdot h + q \cdot x}\right) \cdot c_0$$
 (8)

117 where

119 
$$\chi = \frac{\exp(P_L)}{\exp(P_L) - 1} = \frac{1}{1 - \exp(-P_L)}$$
 (9)

The analytical solution of Eq. (8), associated to the boundary condition

123 
$$c_x(x=0) = c_{x0}$$
 (10)

- where  $c_{x0}$  is the initial groundwater contaminant concentration that comes from upstream of the
- landfill, is given by:

128 
$$c_x = c_{x0} + (c_0 - c_{x0}) - (c_0 - c_{x0}) \cdot \left(\frac{q_{x0} \cdot h}{q_{x0} \cdot h + q \cdot x}\right)^{\alpha}$$
 (11)

Defining the relative concentration, RC, as follows:

132 
$$RC = \frac{c_x - c_{x0}}{c_0 - c_{x0}},$$
 (12)

the analytical solution can also be expressed in the following dimensionless form:

$$136 \qquad RC = 1 - \left(\frac{\eta}{\eta + X}\right)^{\chi} \tag{13}$$

where  $X = x/\ell$  is the dimensionless distance beneath the landfill, with reference to the landfill length

in the direction of the groundwater flow,  $\ell$ , and

140

$$141 \qquad \eta = \frac{q_{x0} \cdot h}{q \cdot \ell} \,. \tag{14}$$

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The parameter  $\eta$  represents the ratio of the horizontal-to-vertical volumetric flow rates. The relative

concentration at a given point in the aquifer beneath the landfill is a decreasing function of  $\eta$ ; i.e.,

when  $\eta \to \infty$ , the relative concentration tends to zero, which corresponds to "perfect flushing", as

shown in Figure 4. The influence of the Peclet number of the barrier on the relative concentration is

limited to values of P<sub>L</sub> lower than 4.

When  $P_L > 4$ , the vertical solute mass flux is dominated by advection, and can be expressed as follows:

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$$\mathbf{J}_{s} = \mathbf{q} \cdot \mathbf{c}_{0}. \tag{15}$$

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Under such conditions, the relative concentration no longer depends on P<sub>L</sub>, and can be expressed as

153 follows:

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$$RC = \frac{X}{\eta + X}.$$
 (16)

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When the vertical volumetric flux is nil (i.e. q = 0), the Peclet number is also nil and the vertical solute

mass flux is given by:

$$\mathbf{J}_{s} = \Lambda \cdot (\mathbf{c}_{0} - \mathbf{c}_{b}). \tag{17}$$

162 Under such conditions, the relative concentration that is obtained from the solute mass balance within

the aquifer can be expressed as follows:

$$165 RC = 1 - \exp\left(-\frac{X}{\eta_D}\right) (18)$$

167 where

$$\eta_{\rm D} = \frac{q_{\rm x0} \cdot h}{\Lambda \cdot \ell} \,. \tag{19}$$

## 3. POLLUTANT CONCENTRATION IN THICK AQUIFERS

When the thickness of the aquifer is not limited to a few meters, the variation in pollutant concentration in the vertical direction becomes significant, and a two-dimensional geometry needs to be taken into account. Therefore, the pollutant mass balance, within a non-deformable aquifer, can be expressed as follows:

$$\mathbf{177} \qquad \mathbf{n_{aq}} \frac{\partial \mathbf{c}}{\partial t} = \mathbf{n_{aq}} \frac{\partial}{\partial \mathbf{x}} \left( \mathbf{D_{h,x}} \frac{\partial \mathbf{c}}{\partial \mathbf{x}} \right) + \mathbf{n_{aq}} \frac{\partial}{\partial \mathbf{y}} \left( \mathbf{D_{h,y}} \frac{\partial \mathbf{c}}{\partial \mathbf{y}} \right) - \frac{\partial}{\partial \mathbf{x}} \left( \mathbf{q_x} \cdot \mathbf{c} \right) - \frac{\partial}{\partial \mathbf{y}} \left( \mathbf{q_y} \cdot \mathbf{c} \right)$$
 (20)

where  $n_{aq}$  is the aquifer porosity, c = c(x,y) is the pollutant concentration within the aquifer, x is the horizontal distance beneath the landfill, y is the vertical distance from the top of the aquifer,  $D_{h,x}$  and  $D_{h,y}$  are the horizontal and vertical hydrodynamic dispersion/diffusion coefficients in the aquifer, respectively, and  $q_x$  and  $q_y$  are the horizontal and vertical components of the groundwater volumetric flux in the aquifer, respectively. Under steady-state conditions, and assuming pure advection as the dominant transport mechanism in the horizontal direction, the mass balance is represented by a parabolic partial differential equation (Rubin and Buddemeier, 1996; Charbeneau, 2000):

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$$\frac{\partial}{\partial x} (q_x \cdot c) = n_{aq} \frac{\partial}{\partial y} \left( D_{h,y} \frac{\partial c}{\partial y} \right) - \frac{\partial}{\partial y} (q_y \cdot c).$$
 (21)

In an aquifer of thickness h with an impermeable layer at the bottom, the vertical volumetric flux  $q_y$  can be assumed to vary linearly with the depth from the vertical infiltration value (i.e. q) at y = 0 to a null value at y = h (Charbeneau et al., 1995):

$$\mathbf{193} \qquad \mathbf{q}_{\mathbf{y}} = \mathbf{q} \cdot \left(1 - \frac{\mathbf{y}}{\mathbf{h}}\right). \tag{22}$$

195 In order to preserve the volumetric balance within the aquifer

196

197 
$$\frac{\partial q_x}{\partial x} + \frac{\partial q_y}{\partial y} = 0, \qquad (23)$$

198

the horizontal volumetric flux must also vary linearly along the horizontal direction, as in Eq. 2:

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201 
$$q_x = q_{x0} + \frac{q}{h} \cdot x$$
. (24)

202

- 203 If the horizontal volumetric flux in the aquifer is appreciably greater than the vertical volumetric flux,
- then the transverse mechanical dispersion can be assumed to be dominant relative to molecular
- 205 diffusion and the longitudinal mechanical dispersion in the vertical direction (Rubin and Buddemeier,
- 206 1996). As a result, the coefficient  $D_{h,y}$  can be calculated as follows:

207

$$D_{h,v} = \alpha_T \cdot v_{x0} \tag{25}$$

209

- where  $\alpha_T$  is the transverse dispersivity within the aquifer and  $v_{x0} = q_{x0}/n_{aq}$  is the horizontal seepage
- velocity of groundwater upstream from the landfill.
- The pollutant mass balance can be expressed, using Eqs. 22, 24 and 25, as follows:

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214 
$$q_x \frac{\partial c}{\partial x} = \alpha_T \cdot q_{x0} \cdot \frac{\partial^2 c}{\partial y^2} - q_y \frac{\partial c}{\partial y}.$$
 (26)

The boundary condition associated with Eq. 26 at the top of the aquifer (i.e. at y = 0) is obtained by imposing continuity between the vertical solute flux coming from the landfill and the vertical solute flux entering the aquifer:

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221

- 222 If the bottom of the aquifer is constituted by an impermeable layer, the boundary condition at y = h
- is given by

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$$\frac{\partial c}{\partial y} = 0 \quad \text{at } y = h.$$
 (28)

226

- The following initial condition is then sufficient to formulate the mathematical problem pertaining to
- 228 the mass balance (i.e. Eq. 26):

229

230 
$$c = c_{x0}$$
 at  $x = 0$ . (29)

- A numerical solution to this problem can be obtained by adopting a step-by-step calculation
- procedure, in which a discretization based on centered finite differences in direction y and the forward
- Euler method are used to integrate with respect to variable x (see Appendix 1).
- However, an analytical solution can be found for cases in which the aquifer thickness is very large,
- by assuming a semi-infinite aquifer and neglecting vertical advection in comparison to vertical
- transverse mechanical dispersion in the mass balance (but not in the boundary condition at y = 0).
- Since  $h \to \infty$  for a semi-infinite aquifer, the horizontal volumetric flux given by Eq. 24 can be
- assumed constant (i.e.  $q_x = q_{x0}$ ) and the mass balance can be expressed as follows:

241 
$$\frac{\partial c}{\partial x} = \alpha_T \cdot \frac{\partial^2 c}{\partial y^2}$$
. (30)

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- An analytical solution to Eq. 30, associated with the boundary condition given by Eq. 27 and the
- initial condition given by Eq. 29, can be derived from the set of solutions provided by Carslaw and
- Jaeger (1959) and Crank (1975) for heat and diffusion problems as follows:

246

$$c(x,y) = c_{x0} + (c_0 - c_{x0}) \cdot \left[ \operatorname{erfc} \left( \frac{y}{2 \cdot \sqrt{\alpha_T \cdot x}} \right) - \exp\left( \gamma \cdot y + \gamma^2 \cdot \alpha_T \cdot x \right) \cdot \operatorname{erfc} \left( \frac{y}{2 \cdot \sqrt{\alpha_T \cdot x}} + \gamma \cdot \sqrt{\alpha_T \cdot x} \right) \right]$$

$$(31)$$

248

249 where

250

$$251 \qquad \gamma = \frac{q}{\alpha_{\rm T} \cdot q_{\rm x0}} \cdot \left[ \frac{\exp(P_{\rm L})}{\exp(P_{\rm L}) - 1} \right]. \tag{32}$$

252

253 The same analytical solution can also be expressed in the following dimensionless form:

254

255 
$$RC = \operatorname{erfc}\left(\frac{Y}{2 \cdot \sqrt{X}}\right) - \exp\left(\Gamma \cdot Y + \Gamma^2 \cdot X\right) \cdot \operatorname{erfc}\left(\frac{Y}{2 \cdot \sqrt{X}} + \Gamma \cdot \sqrt{X}\right)$$
 (33)

256

257 where

259 
$$RC = \frac{c - c_{x0}}{c_0 - c_{x0}}$$
 (34a)

$$260 X = \frac{X}{\ell} (34b)$$

$$Y = \frac{y}{\sqrt{\alpha_T \cdot \ell}}$$
 (34c)

$$\Gamma = \gamma \cdot \sqrt{\alpha_{\rm T} \cdot \ell} = \frac{\chi}{\eta'}$$
 (34d)

263 
$$\eta' = \frac{q_{x0} \cdot \sqrt{\alpha_T \cdot \ell}}{q \cdot \ell}. \tag{34e}$$

When the Peclet number is higher than 4 (i.e.  $P_L > 4$ ), the vertical transport is dominated by advection,

 $\chi$  tends to unity and the parameter  $\Gamma$  reduces to:

$$\Gamma = \frac{\mathbf{q} \cdot \ell}{\mathbf{q}_{x0} \cdot \sqrt{\alpha_{T} \cdot \ell}} = \frac{1}{\eta'}.$$
 (35)

270 If the vertical volumetric flux is nil (i.e. q = 0), the vertical transport is purely diffusive, and the

271 parameter  $\Gamma$  is given by:

$$\Gamma = \frac{1}{\eta'_{D}}.$$
 (36a)

where

277 
$$\eta'_{D} = \frac{q_{x0} \cdot \sqrt{\alpha_{T} \cdot \ell}}{\Lambda \cdot \ell}.$$
 (36b)

If a threshold value of the relative concentration, RC<sub>lim</sub>, is assumed in the analysis of the pollutant migration from the landfill, the thickness of a pollutant plume in the groundwater can be derived through Eq. 33 by finding the depth at which  $RC = RC_{lim}$ . As an example, the thickness of the pollutant plume is plotted in Figure 5 for RC<sub>lim</sub> = 0.001, 0.005, 0.01 and  $\Gamma$  = 0.05. The use of the analytical solution given by Eq. 33 allows to avoid the introduction of a semi-empirical formula for the thickness of the pollutant plume, as described in Charbeneau et al. (1995), as well as the calculation of a boundary layer thickness through approximate solutions, as described in Rubin and Buddemeier (1996).The relevance of neglecting the vertical advective transport in Eq. 30 can be appreciated by comparing the relative concentrations, calculated with Eq. 33, to the numerical solution described in Appendix 1. The relative concentrations determined at X = 1 with both the analytical and numerical solutions are shown in Figure 6 as a function of the dimensionless depth,  $Y' = y/h = Y \cdot (\sqrt{\alpha_T \cdot \ell})/h$ , while varying the upstream horizontal groundwater flow,  $q_{x0}$ , from  $1 \cdot 10^{-8}$  to  $1 \cdot 10^{-6}$  m/s for the following set of parameters:  $q = 1.10^{-10}$  m/s,  $\Lambda = 1.10^{-10}$  m/s,  $P_L = 1$ ,  $\ell = 1000$  m,  $\alpha_T = 1$  m, h = 100 m. The analytical solution appears to be in good agreement with the numerical results, at least as long as the ratio between the vertical volumetric flux, q, and the horizontal volumetric flux,  $q_{x0}$ , is less than 1%, i.e.  $q/q_{x0} < 0.01$ . The approximation arising from the assumption of infinite thickness for the aquifer can be appreciated in Figure 7 by comparing the relative concentration that has been calculated at X = 1 with Eq. 33 to the results provided by the numerical solution, varying the aquifer thickness from h = 20 m to h = 100m for the following set of parameters:  $q=1\cdot 10^{-10}$  m/s,  $\Lambda=1\cdot 10^{-10}$  m/s,  $P_L=1,~\ell=1000$  m,  $\alpha_T=1$ m,  $q_{x0} = 1 \cdot 10^{-6}$  m/s. The analytical solution results are sufficiently accurate for h = 100 m when the ratio of the aquifer thickness, h, to the landfill length,  $\ell$ , is greater than 10% (h/ $\ell$  > 0.1). If this condition is not fulfilled, an improvement to the analytical solution given by Eq. 33 can be obtained by reflecting the concentration curve at the bottom impermeable boundary (i.e. at y = h) and

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superimposing the reflected curve onto the original one. Repeating this procedure a number j=r of times, the resulting solution can be expressed as follows:

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$$RC = \sum_{j=1}^{r} \operatorname{erfc}\left(\frac{2 \cdot Y_{aq} \cdot (j-1) + Y}{2 \cdot \sqrt{X}}\right) - \exp\left[\Gamma \cdot (2 \cdot Y_{aq} \cdot (j-1) + Y) + \Gamma^{2} \cdot X\right] \cdot \operatorname{erfc}\left(\frac{2 \cdot Y_{aq} \cdot (j-1) + Y}{2 \cdot \sqrt{X}} + \Gamma \cdot \sqrt{X}\right) + \operatorname{erfc}\left(\frac{2 \cdot Y_{aq} \cdot j - Y}{2 \cdot \sqrt{X}}\right) - \exp\left[\Gamma \cdot (2 \cdot Y_{aq} \cdot j - Y) + \Gamma^{2} \cdot X\right] \cdot \operatorname{erfc}\left(\frac{2 \cdot Y_{aq} \cdot j - Y}{2 \cdot \sqrt{X}} + \Gamma \cdot \sqrt{X}\right)$$

$$(37)$$

308

where  $Y_{aq} = h/\sqrt{\alpha_T \times \ell}$  is the relative depth of the aquifer. When Eq. 37 is used, the analytical solution is also in good agreement with the numerical results obtained for h = 20 m, as shown in Figure 8.

#### 4. APPLICATION EXAMPLES

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The following examples are provided to illustrate how the previously derived steady-state solutions 313 can be employed in order to assess the equivalency and effectiveness of different landfill barriers. 314 Because any realistic analysis should be based on specific data that have been measured by means of 315 316 field and/or laboratory tests, the results of the following examples are only representative of the proposed analysis approach and should therefore not be generalized to analogous barriers that are 317 characterized by different parameter values and/or are exposed to different boundary conditions. 318 Two barriers scenarios are considered herein. The first barrier scenario is a composite barrier 319 comprising a 1.5-mm-thick geomembrane liner (GML) and a 1-m-thick compacted clay liner (CCL), 320 321 which overlies a 3-m-thick attenuation layer (AL). The second barrier scenario is a composite barrier 322 comprising a 1.5-mm-thick geomembrane liner (GML) and a 10-mm-thick geosynthetic clay liner (GCL), which overlies a 4-m-thick attenuation layer (AL) (Figure 9). The two barriers are therefore 323 324 characterized by approximately the same total thickness (i.e.  $L \cong 4$  m). 325 The height of the ponded leachate in the leachate collection and removal system (LCRS), h<sub>D</sub>, is assumed to be equal to 0.5 m, which is the minimum thickness of the LCRS that is required by 326 European Directive 1999/31/EC, and the hydraulic head at the bottom of the barrier, h<sub>b</sub>, is assumed 327 to be equal to 1.5 m (Figure 9). As a result, the difference in the hydraulic head between the top of 328 the mineral layers and the bottom of the attenuation layer,  $\Delta h$ , is equal to 3 m for the barrier with the 329 CCL and 3.01 m for the barrier with the GCL. The physical, hydraulic and transport parameters that 330 331 have been assigned to the geomembrane and the mineral layers are reported in Figure 9 and in Table 1. 332 The CCL is hypothesized to be characterized by an average value of the hydraulic conductivity that 333 334 corresponds to the maximum value that is permitted by the European and USA regulations, i.e. k =  $1\cdot 10^{-9}$  m/s. The porosity, n, and tortuosity factor,  $\tau_a$ , values have been estimated from the data on the 335 336 kaolinite specimens that were tested by Shackelford and Daniel (1991a,b).

The GCL hydraulic conductivity and porosity values are derived from the results of the laboratory test conducted by Puma et al. (2015) with an aggressive permeant solution of 0.25 M of CaCl<sub>2</sub> under an effective confining stress of 70 kPa. These selected values take into account the increase in hydraulic conductivity and the reduction in void ratio that are induced by a long-term permeation with an aqueous solution having a high salt concentration. The GCL tortuosity factor is derived from the data on the sodium bentonite specimen tested by Dominijanni et al. (2013), neglecting the solute restriction effect that is related to chemico-osmotic phenomena, which represents a conservative assumption. Typical parameter values of a silty soil have been selected for the AL (Manassero et al., 2000; Rowe and Brachman, 2004; Rowe et al., 2004). The analysis is developed for *toluene* (C<sub>6</sub>H<sub>5</sub>-CH<sub>3</sub>), which is a common component of municipal solid waste landfill leachates.

The leakage through composite barriers that include the GML has been calculated as the product of the number of holes per unit area in the GM, n<sub>h</sub>, and the leakage rate through a single hole that coincides with a wrinkle, which, assuming no interaction between adjacent wrinkles, can be written

as follows (Rowe, 1998):

352 
$$Q = 2 \cdot L_{w} \left[ k_{eq} \cdot b + (k \cdot L \cdot \Theta)^{0.5} \right] \frac{\Delta h}{L}$$
 (38)

where  $L_w$  is the length of the wrinkle,  $k_{eq}$  is the hydraulic conductivity the underlying mineral barrier, 2b is the width of the wrinkle, L is the thickness of the underlying mineral layer (or the total thickness of the underlying mineral barrier),  $\Theta$  is the transmissivity of the interface between the GML and the underlying soil, and  $\Delta h$  is the hydraulic head loss across the barrier system.

The equivalent hydraulic conductivity, k<sub>eq</sub>, in Eq. 38 is calculated as the harmonic mean of the hydraulic conductivities of individual layers:

361 
$$k_{eq} = \frac{L}{\sum_{i=1}^{N_1} \frac{L_i}{k_i}}$$
 (39)

where  $k_i$  is the hydraulic conductivity of the i-th layer. The calculation of  $k_{eq}$  also includes the contribution of the attenuation layer that is placed between the engineered barrier system and the underlying aquifer.

The following parameters have been assumed for the calculation of the leakage rate per unit area, q:  $n_h = 1$  hole in a wrinkle per hectare,  $L_w = 3$  m, 2b = 0.2 m,  $\Theta = 4\cdot10^{-8}$  m²/s for the contact between GML and CCL and  $\Theta = 3.5\cdot10^{-11}$  m²/s for the contact between GML and GCL. The value of transmissivity,  $\Theta$ , that has been assigned to the GML - CCL contact represents the average value of the range estimated by Rowe (1998), which varies from  $1.6\cdot10^{-8}$  m²/s to  $1\cdot10^{-7}$  m²/s for this type of composite barrier. Analogously, the value of transmissivity,  $\Theta$ , that has been assigned to the GML - GCL contact represents the average value of the range provided by Harpur et al. (1993), which varies between  $6\cdot10^{-12}$  m²/s and  $2\cdot10^{-10}$  m²/s (Rowe and Brachman, 2004).

The obtained results (Table 2) show that the leakage rate through the composite barrier with GCL (q

= 3.4 lphd) is appreciably lower than the leakage rate through the composite barrier with CCL (q =

9.8 lphd), because of the better contact conditions between the GML and GCL.

The free-solution diffusion coefficient,  $D_0$ , is equal to  $9.7 \cdot 10^{-10}$  m<sup>2</sup>/s for toluene (Yaws, 1995). The average values of the geomembrane partition coefficient and the diffusion coefficient have been assumed equal to  $K_g = 96$  and  $D_g = 0.47 \cdot 10^{-12}$  m<sup>2</sup>/s, respectively (Rowe, 1998), on the basis of the data by Park and Nibras (1993) pertaining to the migration of toluene in aqueous solutions.

The effective diffusion coefficient for the mineral layers has been calculated as the product of the apparent tortuosity factor,  $\tau_a$ , and the free-solution diffusion coefficient (Shackelford and Daniel, 1991a). The longitudinal dispersivity of the mineral layers has been assumed equal to a tenth of the thickness of the barrier layers to calculate the mechanical dispersion coefficients, which are defined

as the product of the longitudinal dispersivity and the seepage velocity (Shackelford and Rowe, 1998; Guyonnet et al., 2001). The hydrodynamic dispersion/diffusion coefficients in the vertical direction,  $D_{hi}$ , have been calculated as the sum of the effective diffusion coefficients and the mechanical dispersion coefficients (Shackelford, 1993). The calculated values of the equivalent diffusivity,  $\Lambda$ , and the Peclet number,  $P_L$ , are reported in Table 2. In the presence of the GML, the Peclet number is lower than 1, thus indicating the dominance of diffusion over advection in the migration process of this organic contaminant, which is able to diffuse through the geomembrane. Similar conclusions were reached by Katsumi et al. (2001), Foose (2010) and Pu et al. (2015, 2016a, 2016b, 2016c, 2019). When the performance of a composite barrier is assessed, the finite service life of the geomembrane needs to be taken into account (Sangam and Rowe, 2002; Rowe, 2005). For example, Rowe (2006) pointed out that the service life of geomembranes is of the order of 15-50 years at temperatures of 50-60 °C. As a result, the volumetric flux through the mineral layers, q, has been calculated conservatively for both of the barriers without taking into account the presence of a geomembrane, in order to assess the barrier performance after geomembrane degradation. Under the assumption of saturated conditions in the liners and the attenuation layer, q has been determined as follows:

401 
$$q = k_{eq} \frac{h_p + L - h_b}{L}$$
 (40)

The CCL barrier is characterized by a value of  $q = 2.91 \cdot 10^{-9}$  m/s, which is significantly lower than the value  $q = 4.39 \cdot 10^{-8}$  m/s that has been found for the GCL barrier. This high value of the volumetric flux of the GCL barrier is related to the degradation of the hydraulic containment ability of GCL, due to the permeation of aggressive aqueous solutions, which has been considered in the selection of the value to assign to the GCL hydraulic conductivity. The Peclet number of the considered example mineral barriers are larger than 10, thus showing that, in the absence of the geomembrane, advection controls the contaminant migration processes.

411 Thin aquifer

If the aquifer beneath the landfill with a length  $\ell$  of 1,000 m is sufficiently thin (h = 3 m), the analytical solution (Eq. 13) can be used to assess the variation of the contaminant concentration in the aquifer along the direction of the groundwater flow.

The values of  $\eta$  and the relative concentration at  $x=\ell$ , which have been calculated assuming that the horizontal groundwater volumetric flux just upstream from the landfill,  $q_{x0}$ , is equal to  $1\cdot 10^{-6}$  m/s (= 31.6 m/yr), are reported in Table 3 for all the considered example barriers. The calculated relative concentrations of toluene in the aquifer are shown as a function of the horizontal distance below the

The composite barrier with the GCL is more effective than the composite barrier with the CCL in reducing the toluene concentrations in the aquifer, even though a conservative value of the GCL hydraulic conductivity, which can be reached after a long-term permeation with an aggressive

aqueous solution, has been assumed. The concentration of toluene increases along the direction of the groundwater flow beneath the landfill. Therefore, the maximum value of contaminant concentration

After the degradation of the geomembrane, the effectiveness of the barriers is reduced significantly and, as a result, the relative concentration below the landfill increases by more than one order of magnitude. Under such conditions, the CCL is more efficient than the GCL, due to the better ability

of the CCL to reduce the contaminant diffusive flux.

is reached at  $x = \ell$ , i.e. just downstream from the landfill.

landfill in Figure 10, for all the considered cases.

Thick aquifer

If the aquifer thickness is not limited to a few meters, the vertical distribution of the contaminant needs to be taken into account by means of the analytical solutions presented by Eqs. 33 or 37, or by means of the numerical solution presented in the appendix. In this case, the contaminant concentration in the aquifer is dependent not only on the horizontal flushing resulting from the groundwater flow,

but also on the vertical dispersion. In this example, the aguifer thickness, h, has been assumed equal 436 to 100 m and the transverse dispersivity within the aquifer,  $\alpha_T$ , has been assumed equal to 1 m, based 437 on the indications of Rowe et al. (2004) in case of availability of high-quality experimental data. 438 The landfill length,  $\ell$ , and the upstream horizontal groundwater volumetric flux,  $q_{x0}$ , have been 439 assumed equal to 1,000 m and  $1 \cdot 10^{-6}$  m/s (= 31.6 m/yr), respectively, in the same way as for the thin 440 aquifer example. 441 The calculated relative concentrations of toluene are shown in Figures 11 as a function of the aquifer 442 depth, y, at the distances x = 100 m, 500 m and 1,000 m (i.e. at X = 0.1, 0.5 and 1) beneath the landfill. 443 444 The contaminant concentration decreases with depth and increases over the horizontal distance. The analytical solution for the barrier constituted by a GCL overlying an attenuation layer is not 445 446 accurate, as the ratio  $q/q_{x0} = 4.4\%$  is greater than 1% (Figure 11d). If a limiting value of the relative concentration is selected, a contaminant plume can be defined within 447 448 the aguifer by means of the available analytical solution. For example, the toluene plumes corresponding to the limiting value of the relative concentration RC<sub>lim</sub> of 0.01% are shown in Figure 449

12 for the two barrier examples with the intact geomembrane.

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#### 5. CONCLUSIONS

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The illustrative examples have shown the application of steady-state analytical solutions for the assessment of the contaminant concentration within an aquifer that is located below a landfill. On the basis of such an analysis, the *equivalence* between landfill barriers which consist of different liners can be established. Moreover, the effectiveness of the barrier in limiting contaminant migration and the related risk to groundwater quality can also be evaluated. With respect to the previous steadystate analysis approaches (Manassero et al., 2000; Guyonnet et. al., 2001; Foose, 2010), the proposed analytical solutions allow the contaminant concentration distribution to be evaluated in the horizonal direction of the groundwater flow, and eventually in the vertical direction for the case of thick aquifers. However, when the ratio between the vertical and horizontal water fluxes, i.e.  $q/q_{x0}$ , is higher than 1%, the proposed analytical solution for thick aguifers is not accurate and should be replaced by the numerical solution that is presented in Appendix 1. The principal benefit of using such solutions is the possibility of conducting an analysis that involves a limited number of parameters and allows the influence of the liner properties (e.g. hydraulic conductivity, thickness, defects) and the field conditions (e.g. aquifer thickness, groundwater velocity) on the final result to be evaluated. Since the assumed boundary conditions are conservative with respect to the evaluation of the contaminant concentration within the aquifer, the proposed analysis can be compared to a Tier-2 risk assessment of the ASTM risk-based corrective action (RBCA) standard (ASTM, 2015) for a polluted site. A possible interesting development of the proposed steady-state analysis method is the application within a probabilistic approach in which the boundary conditions and the model parameters (such as the leachate contaminant concentration, the hydraulic conductivity of the mineral layers and the number, size and location of the geomembrane defects) take on a random nature. In fact, the obtained analytical solutions may be implemented in a Monte Carlo algorithm to relate the pollutant concentration in the aguifer not only to the most representative values of the involved model parameters, but also to their variance.

### 478 **APPENDIX** 1

The pollutant mass balance given by Eq. 26 can be expressed in the following dimensionless form:

480

481 
$$\frac{\partial RC}{\partial X} = d \cdot \frac{\partial^2 RC}{\partial Y'^2} - a \frac{\partial RC}{\partial Y'}$$
 (A1)

482

483 where

484

$$485 Y' = y/h (A2)$$

486 
$$d = \frac{\alpha_{T} \cdot q_{x0} \cdot \ell}{\left(q_{x0} + \frac{q \cdot \ell}{h} \cdot X\right) \cdot h^{2}},$$
 (A3)

487 
$$a = \frac{q \cdot (1 - Y') \cdot \ell}{\left(q_{x0} + \frac{q \cdot \ell}{h} \cdot X\right) \cdot h} .$$
 (A4)

488

- The Y' variable can be discretized into  $N_{Y'} + 1$  nodes, which are numbered from 0 to  $N_{Y'}$  and are
- separated by equal intervals of  $\Delta Y' = 1/N_{Y'}$ . Similarly, the X variable can be discretized into  $N_X + 1$
- nodes, which are numbered from 0 to  $N_X$  and are separated by equal intervals  $\Delta X = 1/N_X$ .
- 492 Using a centered finite difference to approximate the derivatives in variable Y' and the Euler forward
- method to integrate with respect to the variable X, the following step-by-step calculation procedure
- 494 is found:

495

496 
$$RC_{i}^{j+1} = RC_{i}^{j} + \left(d^{j} \cdot \frac{RC_{i+1}^{j} - 2 \cdot RC_{i}^{j} + RC_{i-1}^{j}}{\Delta Y'^{2}} - a_{i}^{j} \cdot \frac{RC_{i+1}^{j} - RC_{i-1}^{j}}{2 \cdot \Delta Y'}\right) \cdot \Delta Y'$$
(A5)

497

498 where

$$RC_{i}^{j+1} = RC(X_{j+1}, Y'_{i})$$

$$RC_{i}^{j} = RC(X_{j}, Y'_{i})$$

$$RC_{i+1}^{j} = RC(X_{j}, Y'_{i+1})$$

$$RC_{i-1}^{j} = RC(X_{j}, Y'_{i-1})$$

$$d^{j} = d(X_{j})$$

$$a_{i}^{j} = a(X_{j}, Y'_{i}).$$

In order to apply Eq. A5 to all the nodes of the Y' variable, two fictitious nodes, which are numbered as -1 and  $N_{Y'} + 1$  and are located outside the grid extremes, have to be introduced. The relative concentrations at these fictitious nodes are determined from the boundary conditions. The boundary condition that is obtained at the top of the aquifer by imposing flux continuity is given by Eq. 27, which can be discretized as follows:

508 
$$G \cdot RC_0^j - \frac{RC_{1}^j - RC_{-1}^j}{2 \cdot \Delta Y'} = G$$
 (A6)

510 where

512 
$$G = \frac{q}{q_{x0}} \cdot \frac{\exp(P_L)}{\exp(P_L) - 1} \cdot \frac{h}{\alpha_T}.$$
 (A7)

Similarly, at the bottom of the aquifer, Eq. 28 can be discretized as follows:

516 
$$\frac{RC_{N_{Y'}+1}^{j} - RC_{N_{Y'}-1}^{j}}{2 \cdot \Lambda Y'} = 0.$$
 (A8)

The resulting step-by-step calculation procedure is:

Initial condition

For i varying from 0 to  $N_{\gamma'}$ 

$$RC_i^0 = 0$$

end

520

521

Integration in X

For j varying from 0 to  $N_x - 1$ 

$$RC_{-1}^{j} = RC_{1}^{j} - G \cdot 2 \cdot \Delta Y' \cdot RC_{0}^{j} + G \cdot 2 \cdot \Delta Y'$$

$$RC^j_{N_{\mathbf{Y}'}+1} = RC^j_{N_{\mathbf{Y}'}-1}$$

For i variying from 0 to  $N_{Y'}$ 

$$RC_{i}^{j+l} = RC_{i}^{j} + \left(d_{i}^{j} \cdot \frac{RC_{i+l}^{j} - 2 \cdot RC_{i}^{j} + RC_{i-l}^{j}}{\Delta {Y'}^{2}} - a_{i}^{j} \cdot \frac{RC_{i+l}^{j} - RC_{i-l}^{j}}{2 \cdot \Delta {Y'}}\right) \cdot \Delta X$$

end

end

In order to obtain a stable solution, the following condition has to be respected in the choice of the

interval of integration  $\Delta X$  (Sewell, 2005):

526 
$$\Delta X \leq \frac{\Delta Y^{\prime 2}}{2 \cdot d_{\text{max}}} = \frac{\Delta Y^{\prime 2} \cdot h^2}{2 \cdot \alpha_T \cdot \ell}.$$
 (A9)

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	a geosynthetic clay liner (GCL).
10	Relative concentration of toluene in a thin aquifer (h = 3 m, $q_{x0} = 1 \cdot 10^{-6}$ m/s)
	beneath the landfill ( $\ell=1000$ m).
	Relative concentration of toluene as a function of the relative depth, $Y' = y/h$ ,
11	at $X = 0.1$ , 0.5 and 1 for the following barriers: (a) $GML + CCL + AL$ ; (b)
	GML + GCL + AL; (c) CCL + AL (degraded geomembrane); (d) GCL + AL
	(degraded geomembrane).
12	Toluene plume beneath the landfill for $RC_{lim} = 0.0001 (0.01\%)$ .