

This discussion paper is/has been under review for the journal Drinking Water Engineering and Science (DWES). Please refer to the corresponding final paper in DWES if available.

Groundwater contamination due to lead (Pb) migrating from Richmond municipal landfill into Matsheumhlope aquifer: evaluation of a model using field observations

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Received: 12 April 2010 – Accepted: 7 September 2010 – Published: 18 October 2010

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Published by Copernicus Publications on behalf of the Delft University of Technology.

DWESD

3, 251–269, 2010

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Abstract

Disposal of solid waste in landfills is an economic option for many municipalities in developing countries where alternatives like incineration and composting are costly. However, groundwater pollution from the leachate generated within the landfill and migrating through the bottom liner material into the underlying groundwater aquifers remains a major public health concern. In our study, we evaluated the application of a mathematical model to determine the aerial extent of unacceptable groundwater contamination due to lead migrating from the Richmond landfill leachate into the underlying Matsheumhlope unconfined aquifer. A one-dimensional advection-dispersion model was applied to predict the down-gradient migration of lead into the aquifer. Linear sorption and first-order decay were considered as the dominant contaminant sink mechanisms for lead. Lead concentrations in the monitoring wells at the landfill site were used as the source term. The lead migration from the landfill was determined by water quality sampling from boreholes situated down-gradient of the landfill. The model simulations gave a good fit of the field results. The safe distance for potable water abstraction was determined to be 400 m, and the model simulations showed that the aerial extent of the pollution will increase with time. The model is most sensitive to the partition coefficient, hydraulic conductivity and longitudinal dispersivity, whilst it exhibits no sensitivity to the lead decay coefficient.

1 Introduction

Sanitary landfills remain the most cost effective option for disposal of solid waste (Islam and Singhal, 2002; El-Fadel et al., 1997) especially for many municipalities in developing countries where the high costs of alternatives such as incineration and composting are prohibitive. However, groundwater pollution from the leachate generated within the landfill and migrating through the liner material into the underlying aquifers remains a major public health concern (Fatta et al., 1999; Nyengera, 2005).

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One area of concern is the Richmond residential suburb in the City of Bulawayo, Zimbabwe. A sanitary landfill is located in the suburb, and overlies a shallow unconfined aquifer, the Matsheumhlope aquifer. The aquifer is a source of potable water for the residents of Richmond and the aquifer is also a potential source of water for the entire City, which is in a drought-prone region of the country.

Nyengera (2004) concluded that the chemical quality of groundwater in the Richmond area with respect to heavy metals and some chemical parameters far exceeds the limits and guideline values for drinking water. The average concentration of mercury and lead were 0.04 mg/l and 0.22 mg/l respectively. Overall, the level of contamination in the boreholes in the Richmond area was very high compared to the average of the other boreholes that were tested in the City. Landfill leachate from the Richmond Landfill migrating into the groundwater aquifer was identified as the possible cause for the high pollution levels in the groundwater.

While the previous study highlighted the current levels of pollution, it did not establish the relation between the leachate occurrence and transport from the landfill through the aquifer, an aspect which this study seeks to address. The long term impact of landfill leachate on groundwater quality will depend on the quality and quantity of the leachate, performance of the liner material and the site geo-hydrology. Decision support systems are then required to predict the subsurface transport of the contaminant in both space and time, and establish the aerial extent of the pollution. Such information is important in the design and scheduling of remediation strategies. The objective of this study was therefore to track the transport of the contaminant in both space and time from the landfill through the porous medium by application of mathematical modeling.

2 Theoretical considerations and model development

Mathematical modelling has been widely applied as a decision support tool to simulate processes governing leachate generation and transport. These models have been successful more in estimating leachate quantity and transport, than its composition

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because of the inherent difficulties associated with estimating model parameters that can adequately describe the complex biological, chemical and physical processes occurring in landfills (El-Fadel et al., 1997). Considerable success has been reported in modelling leakage of leachate through the liner material (Rowe, 1987, 1989) and transport in the subsurface (Ostendorf et al., 1984; Islam and Singhal, 2002) under various field conditions. Modelling of subsurface contaminant transport has also been successfully applied in other fields such as suspended solids and bacterial transport in silted river beds (Mutsvangwa et al., 2006, 2005), bacterial and virus transport in groundwater (Vilker and Burger, 1981; Matthess et al., 1981; Harvey and Garabedian, 1991).

In our study we demonstrate the application of mathematical modelling to determine the aerial extent of unacceptable groundwater contamination due to migration of lead from landfill leachate into a groundwater aquifer.

Lead is an ideal chemical to employ as a tracer chemical for subsurface contamination because it is prevalent in most municipal landfill leachate (Fatta et al., 1999; Ehrig, 1983). Its non-biodegradable and accumulative characteristics leave a long record of contamination in the soil and groundwater. Furthermore, its isotopic ratio can be used as a “fingerprint” to identify anthropogenic sources. It is therefore for these reasons that lead has been widely used as a tracer for leachate pollution in soil and groundwater (Vilomet et al., 2003), and we also employed it in this study. Moreover, lead has adverse health effects to humans such as mental and reproductive impairment, high blood pressure and kidney problems (Manhan, 1991; Ho et al., 2002; Benjamin et al., 1982) and thus is a major public health concern.

The primary transport mechanisms of contaminants in porous media are advection and dispersion (Rowe, 1987; El-Fadel et al., 1997; Ogata and Banks, 1961; Freeze and Cherry, 1979). Contaminant sinks are biochemical and physicochemical processes which include exchange reactions, precipitation and microbial reactions.

In this study, we consider linear sorption (Freeze and Cherry, 1979; Rowe, 1989) and first-order decay as the dominant contaminant sinks for lead. Precipitation may

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occur in soil in the presence of carbonate if the soluble concentrations of the lead exceed 4 mg/L (Rickard and Nriagu, 1978). However for the field conditions considered in this paper, lead concentrations are below 1 mg/L, making precipitation a less important sink mechanism. For a homogeneous and isotropic aquifer and neglecting mild differential density effects which tend to establish vertical concentration gradients within a contaminant plume (Wilson and Miller, 1978) enables the consideration of a simple one-dimensional advection-dispersion model. The model describing the lead concentration at time t and horizontal distance x down gradient of the landfill in the direction of advection velocity as subjected to linear sorption and first-order decay can be written as:

$$\frac{\partial}{\partial t}(\theta c) = \frac{\partial}{\partial x} \left(\theta D \frac{\partial c}{\partial x} \right) - \frac{\partial}{\partial x}(qc) - \rho K_p \frac{\partial c}{\partial t} - \alpha \theta c - \beta \rho S \quad (1)$$

Where c is the depth averaged plume concentration; θ is the volumetric moisture content; D is the hydrodynamic dispersion coefficient accounting for both diffusion and mechanical dispersion; q is the Darcy velocity; ρ is the porous media bulk density; K_p is a partition coefficient; and α and β are first-order rate constants associated with the liquid and solid phases of the soil respectively.

We further define a retardation factor R as:

$$R = 1 + \rho K_p / \theta \quad (2)$$

and a general decay constant μ given by:

$$\mu = \alpha + \beta \rho K_p / \theta \quad (3)$$

Combining Eqs. (1), (2) and (3) gives:

$$\frac{\partial}{\partial t}(\theta R c) + \theta \mu c = \frac{\partial}{\partial x} \left(\theta D \frac{\partial c}{\partial x} \right) - \frac{\partial}{\partial x}(qc) \quad (4)$$

Substituting for the pore-water velocity v ($= q/\theta$), Eq. (4) reduces to:

$$R \frac{\partial c}{\partial t} + \mu c = D \frac{\partial^2 c}{\partial x^2} - v \frac{\partial c}{\partial x} \quad (5)$$

Equation (5) can be solved subject to specified initial and boundary conditions. The initial condition is given by:

$$c(x, 0) = C_i \quad (6)$$

Equation (6) describes the initial lead concentration values at different locations. The boundary conditions associated with the surface are:

$$c(0, t) = C_o \quad (7)$$

$$\text{and } \frac{\partial c}{\partial x}(\infty, t) = 0 \quad (8)$$

Equation (7) is derived from the consideration that for an active landfill, there is continuous inflow of contaminant which replaces the outflow into the soil. At steady state conditions, the contaminant concentration and flux will tend to a constant maximum value (Rowe, 1987). Equation (8) simply entails that at very large distances the aquifer is unaffected by the contamination leaching from the landfill which we consider to be a plausible boundary condition.

The solution of Eq. (5) subject to Eqs. (6), (7) and (8) can be obtained using Laplace transforms and is given as (Genuchten, 1980):

$$c(x, t) = C_o A(x, t) + B(x, t) \quad (9)$$

where

$$A(x, t) = \frac{1}{2} \exp\left[\frac{(v-u)x}{2D}\right] \operatorname{erfc}\left[\frac{Rx-ut}{2(DRt)^{1/2}}\right] + \frac{1}{2} \exp\left[\frac{(v+u)x}{2D}\right] \operatorname{erfc}\left[\frac{(Rx+ut)}{2(DRt)^{1/2}}\right] \quad (10)$$

$$B(x, t) = -C_i \exp\left(-\frac{\mu t}{R}\right) \left\{ \frac{1}{2} \operatorname{erfc}\left[\frac{(Rx-vt)}{2(DRt)^{1/2}}\right] + \frac{1}{2} \exp\left(\frac{vX}{D}\right) \operatorname{erfc}\left[\frac{(Rx+ut)}{2(DRt)^{1/2}}\right] \right\} + C_i \exp\left(-\frac{\mu t}{R}\right) \quad (11)$$

$$u = v(1 + 4\mu D/v^2)^{1/2} \quad (12)$$

Equations (9), (10), (11) and (12) will be applied to predict one-dimensional lead transport during transient fluid flow subject to linear sorption and first-order decay.

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3 Study site

The study site is the Richmond municipal landfill in urban Bulawayo. The landfill covers a total area of 13 353 m², and is bounded by Richmond and Cowdray Park residential suburbs in the east and west respectively. The landfill is the main disposal site for both industrial and domestic solid waste generated in the city. The topography of the area is the post-African Miocene age. The Matsheumhlope aquifer is largely unconfined with an average thickness of 40 m. The geology is dominated by fractured meta-basalt formations (Weaver, 1992). The effective porosity is 0.05, and average hydraulic conductivity is 0.55 m/day (Rusinga and Taigbenu, 2005). The pattern of groundwater flow generally follows the surface topography, which gives the aquifer an average hydraulic gradient of 0.004. Richmond aquifer is classified as largely homogenous and isotropic (Weaver, 1992; Rusinga and Taigbenu, 2005). The aquifer has a long term sustainable annual yield of $6.1 \times 10^6 \text{ m}^3$ (Rusinga and Taigbenu, 2005) which represents about 10% of the city's annual water demand. Therefore the control of pollution into the aquifer is of strategic importance to the future water supply of the city. Privately owned boreholes in the vicinity of the landfill are currently the most susceptible to the effects of contamination from the leachate plume (see Fig. 1).

The bottom of the landfill is lined with compacted clay and mechanical equipment is used to compact the waste. Upon reaching the landfill bottom, some of the leachate will travel laterally through a system of under-drains into three collection ponds shown in Fig. 1. Because compacted clay is not completely impermeable, some of the leachate will inevitably be transported through the clay barrier into the subsurface (Rowe, 1987, 1989; Lee and Jones, 1994).

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4 Model application to study site

4.1 Field results

Lead concentrations in the subsurface were determined at the landfill site and at various points down gradient of the landfill in the direction of groundwater flow. Samples were collected for analysis from the three monitoring wells at the landfill site (Pond 1, 2, and 3) and from the eight privately-owned boreholes down-gradient of the landfill (B1 to B8) as shown in Fig. 1. In order to fit the sampling exercise into our modelling approach, we adopted the approach of Ostendorf et al. (1984) by distinguishing a *near field* region close to the landfill, where mixing of leachate and groundwater is presumed to occur, and a *far field* region of fully mixed, one-dimensional contaminant flow. Consequently, the three monitoring wells at the landfill site are located in the near field and an average value of the contaminant levels in these wells was used to satisfy Eq. (7). The eight boreholes lie in the far field. Equation (5) predicts the contaminant levels in the far field at different time scales.

Polypropylene samplers with a diameter of 95 mm and a volume of 1000 ml (Fisher Scientific) were used for sampling. Polyethylene containers were used for storing samples during transportation to the laboratory. Dissolved gases, oxidisable or reducible constituents are very unstable and can alter the composition of the sample. A change in the composition of the constituents was retarded by storing the samples at 4 °C and exclusion of light during transportation and conducting the analyses immediately upon arrival at the laboratory. The lead concentrations in each sample were determined spectrophotometrically in accordance with Standard Methods (APHA, 1998).

4.2 Aquifer parameters

Data on the properties of the aquifer was obtained from Rusinga and Taigbenu (1995): hydraulic conductivity (K) of 0.55 m/day; a hydraulic gradient (i) of 0.004 m/m and an effective porosity (θ) of 0.05. Applying Darcy Law yields a value of 0.0022 m/day for the Darcy velocity (q) and a pore water velocity ($v = q/\theta$) of 0.044 m/day.

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4.3 The decay constant (μ)

Natural lead is a mixture of four isotopes, ^{208}Pb (51–53%), ^{206}Pb (23.5–27%), ^{207}Pb (20.5–23%) and ^{204}Pb (1.35–1.5%), and exists in three oxidation states Pb^0 , Pb^{2+} , and Pb^{4+} . The decay coefficient of radiogenic lead (μ) varies with each type of isotope. Faure (1986) reported values of $^{208}\text{Pb} = 4.948 \times 10^{-11}/\text{year}$, $^{207}\text{Pb} = 9.848 \times 10^{-10}/\text{year}$, and $^{206}\text{Pb} = 1.551 \times 10^{-10}/\text{year}$. ^{204}Pb is not radiogenic. We used a weighted decay constant of $2.779 \times 10^{-10}/\text{year}$.

4.4 Hydrodynamic coefficient (D)

The hydrodynamic coefficient (D) is a product of the longitudinal dispersivity (a_L) and the pore water velocity (v) and can be determined using trace experiments (Ogata and Banks, 1961; Mutsvangwa et al., 2006). In the absence of tracer tests, the longitudinal dispersivity can be determined using theoretical models (Gelhar and Axness, 1983) or by fitting model simulations to field results. In our study the hydrodynamic dispersion was determined by fitting model simulations to field results.

4.5 Retardation factor (R)

The retardation factor (R) is a function of the partition coefficient (K_p) and bulk media density (ρ) as shown by Eq. (2). The soil bulk density can be readily determined according to normal geotechnical practice and a value of 2800 kg/m^3 was obtained. K_p is unique for every chemical constituent and depends on the mineralogical composition of the soil and the proportion of other non-mineral constituents and thus can vary significantly from one soil to another for a given chemical. K_p should be determined experimentally using the actual soil of interest for the range of chemical concentration expected in the field (Rowe et al., 1988) or by fitting model simulations to field results. The aquifer formation has been classified as fractured meta-basalt formations (Weaver, 1992), which falls under fine graded sand of less than 0.25 mm diameter.

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Various values of K_p for sand formation depending on pH are reported in literature. Lee et al. (1998) reported values of K_p ranging from 10.8 to 575.8 ml/g and Sheppard et al. (1989) gave values ranging from 19 to 59000 ml/g. In this study K_p was determined by fitting model simulations and field results and value of 115 ml/g was established for the aquifer.

Table 1 provides a summary of the model input parameters together with their maximum and minimum values. We select the minimum and maximum values for the other parameters from the literature to show the range of the possible parameters. The adopted model input parameters, including the calibrated parameters are within this range and we consider them to be plausible.

5 Results and discussion

Nyengera (2005) implicated the Richmond landfill leachate as the primary source of the lead contamination in the aquifer. Therefore model simulations were generated by assuming an initially lead-free aquifer prior to leachate contamination. The simulation was then run for the 22 years that the landfill has been in operation which corresponds to the period that the aquifer has been exposed to the leachate contamination. The results of the simulation are shown in Fig. 2, which shows a good fit of the model simulations to the lead concentration levels obtained from the boreholes down gradient of the landfill. The model simulations show that the distance at which the lead concentration in groundwater is less than 0.01 mg/l (safe distance) is about 400 m. Furthermore, the region of contamination will extend with time as shown in Fig. 3.

5.1 Sensitivity analysis of input parameters

Parameter sensitivity analysis involves a series of tests during which the modeller sets different parameter values to see how a change in a parameter value causes a change in the model results. The minimum and maximum values of the input parameters were

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applied into the model while holding all the parameters at the values determined earlier (see Table 1). The corresponding distance at which the lead concentration = 0.01 mg/L was then determined to establish the range of values that can be obtained for the safe distance when different parameters are at their extreme values. Additionally, the magnitude of the range of values gives an indication of the relative accuracy with which the input parameter value should be determined. Input parameters that were calculated from the other parameters were not considered for the sensitivity analysis.

The above analysis shows that the model is most sensitivity to the partition coefficient, hydraulic gradient, and the hydraulic conductivity in that order. The model is marginally sensitive to the medium porosity and exhibits no sensitivity to the decay coefficient. Therefore the values of the partition coefficient, hydraulic gradient, and the hydraulic conductivity should be determined with high accuracy if done experimentally. The magnitude of variation of the lead decay coefficient due to isotropic composition does not affect the model results and thus it was not necessary to determine the isotropic composition of the lead for this study.

6 Conclusions and recommendations

The down-gradient migration of contaminants from the Richmond Landfill is contaminating the Matsheumhlope aquifer which is a source of water to the City of Bulawayo. The concentration of lead in the nearby boreholes exceeds the recommended value in potable water. The results of the model simulation fit the field results and hence the model gives a good prediction of the transient migration of lead from the landfill into the aquifer. Applying the model to the landfill, and taking into consideration the recommended value of lead in drinking water of 0.01 mg/L (WHO, 2004), the current set-back or safe distance for potable water abstraction should be at least greater than 400 m. The zone of contamination by the lead will increase with time as indicated in the simulations. Therefore, the aerial extent of unacceptable lead pollution due to landfill leachate needs to be reviewed from time to time.

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Extreme caution should be taken in utilising the wells, which are within the influence of the lead contamination from Richmond landfill. Negative health impacts due to high concentration of lead are not documented in Richmond area. However, the possibility of occurrence of negative health impacts associated with leachate contamination is very high and future research should focus in this area.

Only lead has been considered in this study and its presence is above the recommended guideline in drinking water. The presence of other carcinogenic contaminants is highly possible like cadmium which is frequently found with lead and can even be transported further from the landfill than lead. Future investigations should consider other contaminants migrating from the landfill which are polluting the aquifer.

There is need to mitigate against the release and transport of contaminants into the Richmond aquifer through improvements in lining or relocating the waste containment facilities away from potential groundwater source or to have grout curtains.

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Table 1. Summary of model parameters.

Description	Symbol	Units	Value			Reference
			Min	Model	Max	
Hydraulic conductivity	K	m/day	0.10	0.55	2.09	Taigbenu and Rusinga (2005)
Porosity	θ	–	0.02	0.05	0.11	Taigbenu and Rusinga (2005)
Hydraulic gradient	i	m/m	0.001	0.004	0.0185	Taigbenu and Rusinga (2005)
Darcy velocity	q	m/day	0.0001	0.0022	0.2299	Calculation
Fluid velocity	V	m/day	0.0009	0.044	11.495	Calculation
Longitudinal dispersivity	a_L	m	15	185	200	Rowe (1987)
Hydrodynamic dispersion coefficient	D	m ² /day	0.0144	8.14	2299	Calculation
Bulk density of porous media	ρ_b	kg/m ³	N/A	2600	N/A	Experimental
Partition coefficient	K_p	ml/g	10.8	115	59 000	Lee et al. (1998), Sheppard et al. (1989)
Retardation coefficient	R	–	1	7	10 000	Todd (2005)
Decay constant	μ	year ⁻¹	4.95×10^{-11}	2.78×10^{-10}	9.85×10^{-10}	Faure (1986)

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Table 2. Sensitivity analysis of the variables that determine the lead concentration.

Description	Units	Value			Safe distance (m)			
		Min	Model	Max	Min	Model	Max	Range ^a
Hydraulic conductivity	m/day	0.10	0.55	2.09	160	400	860	176%
Porosity	–	0.02	0.05	0.11	418	400	364	14%
Hydraulic gradient	m/m	0.001	0.004	0.0185	187	400	971	197%
Longitudinal dispersivity	m	15	185	200	147	400	412	67%
Partition coefficient	ml/g	10.8	115	1295	952	400	17	234%
Decay constant for lead	year ⁻¹	4.9×10^{-11}	2.7×10^{-10}	9.8×10^{-10}	398	400	398	0%

^a The range is calculated as the difference between the maximum and minimum as a percentage of the model value of 400 m for the safe distance.

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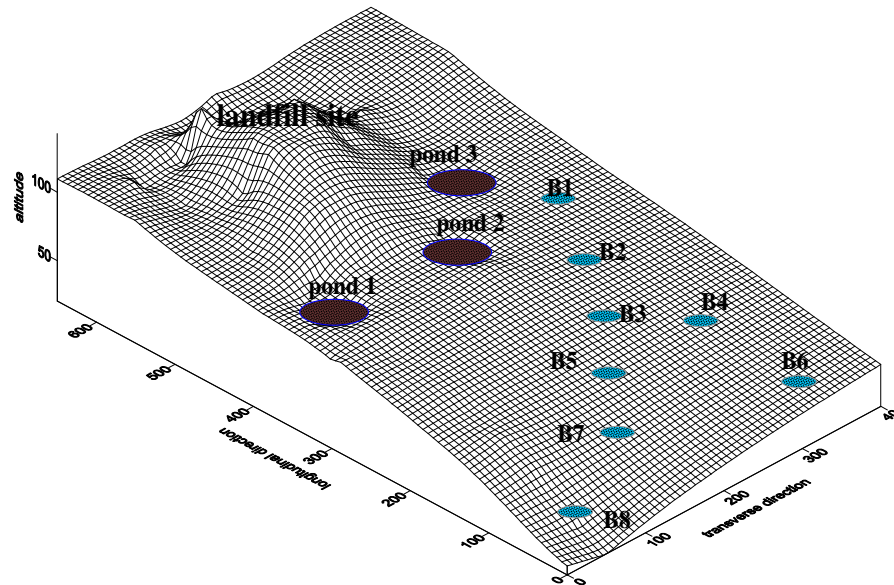


Fig. 1. The study area in 3-D Grid showing the location of the Richmond Landfill, leachate ponds and the boreholes in the Richmond residential area (B1 to B6), which were used as the sampling points. B1 =4 Princess Road; B2 = 10 Cunningham Road; B3 = 32 Cunningham Road; B4 = 1 Brooke road; B5 = 19 Nerine Road; B6 = 6 Erine Road; B7 = 56 Pumula Road; and B8 = 54 Alexander Drive.

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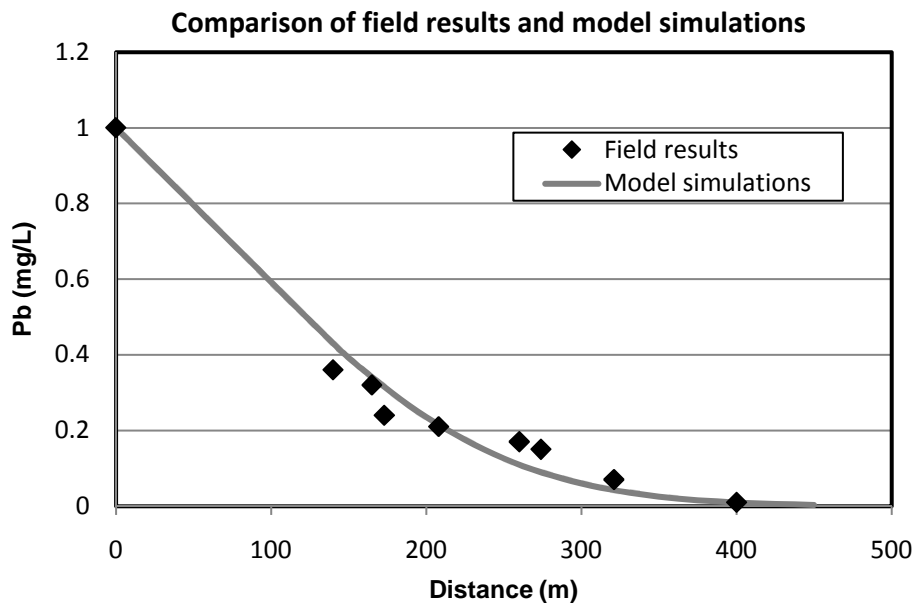


Fig. 2. Fitting model simulations to field results of the lead concentration in the boreholes sampled.

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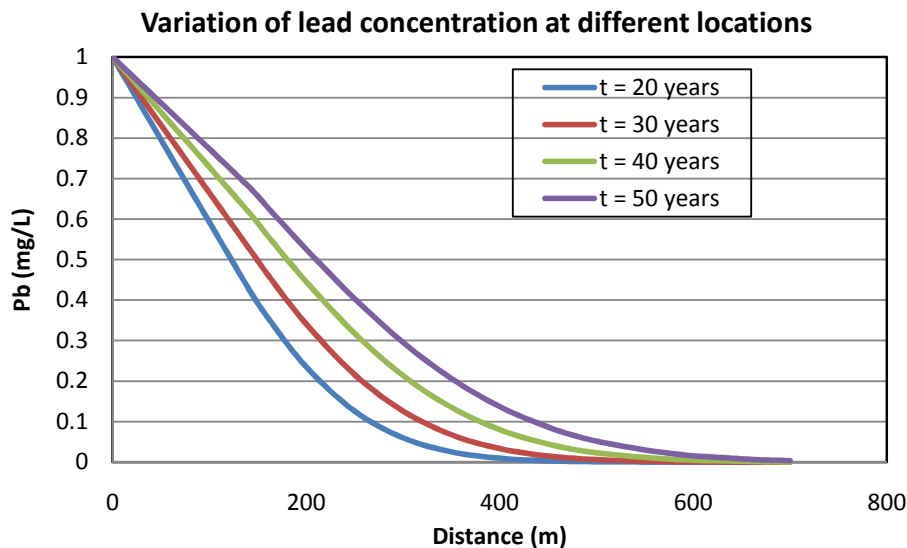


Fig. 3. Variation of lead concentration at different locations with time.

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