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Quantification of Emission Potential of Landfill Waste Bodies Using a Stochastic Leaching Framework

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Key Points:

- Landfill emission potential, the amount of mass that can leach from a waste body, can be quantified by modeling the water and mass balance
- Stochastic travel time based on life expectancy distributions allows for quantification of the emission potential of conservative species
- Emission is driven by net precipitation and long-term precipitation scenarios allow for estimation of future emission dynamics

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract Sanitary engineered landfills require extensive aftercare to safeguard human health and the environment. This involves monitoring emissions like leachate and gas, maintaining cover layers, and managing leachate and gas collection systems. Researchers have explored methods to conclude or extend aftercare. Quantifying emission potential, a key concept integrating various processes influencing emissions, is essential for managing and predicting landfill impacts. In this study we developed a stochastic travel time model based on water life expectancies. The model is used to predict leachate production rates and leachate chloride concentrations from landfill waste bodies. Unknown parameters are quantified by matching model output to measured time series using Bayesian inference. Once parameter distributions have been obtained, we are able to describe the measured long-term leachate dynamics. By analyzing the parameters and evolution of model states, we obtain a deeper understanding of the water and mass balance of the waste bodies. We demonstrate that the model can be used to quantify the chloride emission potential and the estimated values of total chloride mass match data quantified by sampling from the waste body. The results confirm that emissions with leachate are dominated by preferential flow infiltrating from the cover layer. Similar results have been obtained by applying the model to datasets from four different waste bodies, demonstrating that the approach is generally applicable for conservative solutes. Understanding of the water balance of the landfill together with conservative solute leaching is a necessary first step for further evaluating emission of reactive species.

Plain Language Summary Landfills have historically been the primary waste disposal method in Europe, leading to numerous legacy sites requiring extensive aftercare to safeguard human health and the environment. Long-term aftercare of sanitary engineered landfills requires insight in the risk associated with possible emission of contaminants from the landfill waste body. In this research we developed an approach where data measured at the landfill sites are combined with a model in order to quantify the amount of mass that will emit from the waste body over a certain period of time. This is important information for landfill operators and responsible authorities in order to develop approaches for landfill aftercare. Especially because after-care is expected to last for a very long time (up to several centuries).

1. Introduction

Landfills have long been the primary method of waste disposal in Europe, resulting in a large number of legacy landfills that require aftercare to protect human health and the environment. Aftercare typically involves monitoring emissions such as leachate and gas, as well as maintaining the cover layer and collection systems. The European Landfill Directive (EC, 1999) mandates a minimum 30-year aftercare period, but regulatory authorities may choose to shorten or extend this period based on a range of factors. To aid in decision-making, several authors have reviewed different approaches to ending or prolonging landfill aftercare (Barlaz et al., 2002; Laner et al., 2011). Several authors (Butt et al., 2008; Laner et al., 2012) have advocated for using risk-based assessments to evaluate the potential for harm. By exploring these and other solutions, we can work toward reducing the environmental impact of legacy landfills while ensuring the continued protection of public health.

In many cases the aftercare of sanitary engineered landfills consists of post-closure monitoring of emissions (e.g., leachate and gas) and maintenance of the cover layer and leachate and collection systems. Laner et al. (2012) postulate that post-closure care can end once a landfill no longer poses a threat to human health and the environment. Quantitative predictions of future emissions are important in order to assess future threat. Barlaz et al. (2002) advocate the use of technical criteria based on measured time-series of leachate composition, and leachate and gas production rates. One such technical criterion is the presence of barrier systems which require maintenance during the aftercare period. Laner et al. (2011) address the importance of assessing the remaining substance release potentials. The assessment should be site specific and take into account the deposited waste and the relevant boundary conditions that influence the flow of water through the landfill including the performance of the barrier systems. They propose a continuous emission model assuming that the status quo persists (after installation of a cover layer) based on a first order decay rate as proposed by Belevi and baccini (1989). Although Laner et al. (2011) give suggestions how to quantify the remaining substance source term, no experimental data are provided.

A research program aiming to achieve a significant reduction in emissions from Municipal Solid Waste landfills is currently being carried out in the Netherlands (Kattenberg et al., 2013). More information on this program can be found at the website of the research program (Stichting Duurzaam Storten, 2022a, 2022b). In this program different approaches to stabilize waste bodies by irrigation, recirculation and discharge of leachate and aeration of the waste body are being tested. The approaches are tested at full-scale at three different landfills. In order to assess the success of the stabilization measures, site specific Environmental Protection Criteria have been derived (Brand et al., 2016; Dijkstra et al., 2018). These criteria are defined to be the maximum allowable concentration of contaminant in the drainage system below the waste body, which will not lead to a concentration in the groundwater 20 m downstream of the landfill which damages human health or the ecosystem. The underlying modeling approach assumes that source term for all compounds in the drainage system remain constant over the complete evaluation period of 500 years.

In all the papers cited above, there is a common agreement that it is important to have a quantitative understanding of the source term controlling emissions of the contaminants present in the waste body. In this paper we would like to propose some definitions in order to clarify different approaches to quantify the source term.

The total mass of different compounds can be measured in the laboratory from samples taken from the field using destructive analytical techniques. Because contaminants can be bound in solids, total mass can lead to a significant overestimation of leachable mass. Leachable mass can be quantified in the laboratory using different types of leaching methods (Kosson et al., 2002; van der Sloot et al., 2017). However, characterization of heterogeneous landfill waste bodies using sampling and laboratory analysis requires a large amount of samples because of the inherent uncertainty caused by spatial variability (Sormunen et al., 2008a, 2008b). In addition to the spatial variability it is also important to realize that waste bodies most likely contain zones or pockets which isolate volumes of waste from mobile water, such as waste stored in a closed plastic bag. The presence of dead zones implies that not all contamination present in the waste body will be released from the waste body. Consequently, laboratory techniques to assess the source term of contaminants may overestimate the amounts that can be released under field conditions.

In this paper we introduce the term emission potential in order to describe the amount of mass that can be released from the waste body. The emission potential is the result of all processes involved in causing emissions of compounds from waste bodies. It is related to the multi-physical coupling between fluid flow, solute transport, bio geochemical transformations, waste body settlements and many more. The emission potential is the source term in a modeling framework that is able to describe the leachate flux and leachate concentration as a function of time. For conservative solutes we hypothesize that the emission potential can be quantified by fitting models to measured time series. Finally, using these models allows us to quantify the emission from the waste body under different scenarios, for example, over a time period of 500 years. Understanding the water balance and mass balance behavior of conservative solutes is a necessary first step for evaluating the emissions of reactive compounds.

Fellner and Brunner (2010) give an overview of modeling approaches available in the literature for quantifying leachate production. They show that preferential flow is a dominant process in waste bodies and needs to be incorporated in models in order to describe landfill leachate production dynamics (Fellner & Brunner, 2010; Uguccioni & Zeiss, 1997). In order to describe the heterogeneous flow and transport through waste bodies, several Lagrangian based travel time models have been developed (Malmström et al., 2004; Rosqvist & Destouni, 2000; Zacharof & Butler, 2004a, 2004b). Lagrangian modeling of water flow and solute transport in catchment systems has seen significant progress since then (Benettin & Bertuzzo, 2018; Benettin et al., 2015; Benettin et al., 2015, 2015, 2017; Harman, 2015; Harman, 2015, 2015; Hrachowitz et al., 2016; Rinaldo et al., 2015). The advantage of these Lagrangian approaches is that is allows for describing water flow and solute transport in large scale systems where heterogeneity is captured through probability distributions.

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Given the large heterogeneity present in landfills we have developed a stochastic Lagrangian travel time modeling framework to simulate landfill water and mass balances in order to quantify the emission potential of waste bodies. The parameters in the model are calibrated using time series measurements of leachate volumes pumped from the drainage system and bi-weekly chemical analyses of chloride concentrations in pumped leachate. Measuring pumped leachate volumes and leachate quality is standard procedure for landfill operators in the Netherlands and as such obtaining these time series is much easier and cheaper than taking samples from the landfill for analysis in the laboratory. The source term in the model after calibration is considered to be the emission potential. Long-term extrapolations using the calibrated model provide insight how the emission potential impacts future leachate quality. The generic feasibility and applicability of the proposed concept for quantifying the water balance and the leaching of conservative solutes is demonstrated using data from four different waste bodies.

2. Theory and Methods

In order to develop the model equations we hypothesize that the chloride concentration in (pumped) leachate is mainly controlled by dilution of highly concentrated base flow from the waste body with infiltrating water originating from rainfall. We assume that the waste body is a causal system for leachate production and solute transport. This implies that the output of the system at any given time depends only on the input and the system's past behavior and consequently, the flow of water and transport of solutes through the waste body can be modeled using travel time probability distributions. In this paper we assume that the travel time probability density functions (pdf) are constant in time. The approach we follow is similar to the one described by Benettin et al. (2015).

If we follow a water parcel that enters the waste body at time t_{in} and exits the waste body at time t_{ex} then the total time in the waste body ($T_T = t_{ex} - t_{in}$) at any moment in time can be characterized by its age (or residence time) indicated as T_R and the time it will still remain in the waste body before it exits the waste body (its life expectancy) indicated as T_E. Residence time and life expectancy are related to the total travel time T_T by:

$$T_T = T_R + T_E. \tag{1}$$

Each day as the water parcel moves through the waste body, its residence time increases with 1 day and its life expectancy decreases with one day which can be written as:

$$\frac{\mathrm{d}T_R}{\mathrm{d}t} = 1, \; \frac{\mathrm{d}T_E}{\mathrm{d}t} = -1. \tag{2}$$

Equation 2 can be seen as a celerity with unit value, where the sign determines which property is described.

2.1. Water Balance

The upper boundary of the landfill is its surface where water can enter as rainfall and leaves as evapotranspiration. The lower boundary consists of the drainage system where water is pumped out of the landfill as leachate. To simplify the problem we assume that we can model the landfill as a one-dimensional 2-layered column, where the first layer represents a cover layer and the second layer is the waste body.

The water storage in the landfill is defined as:

$$S_w = \frac{V_w}{A_{lf}} = S_{cl} + S_{wb},$$

in which V_w is the is the volume of water per unit area and A_{lf} is the surface area of the landfill and S_{cl} and S_{wb} are the water storage in the cover layer and waste body respectively.

2.1.1. Water Balance of the Cover Layer

The water balance of the cover layer links water entering the landfill as rain and leaving the landfill as evapotranspiration to the amount of water infiltrating in to the waste body:

$$\frac{\mathrm{d}S_{cl}(t)}{\mathrm{d}t} = q_{rf}(t) - q_{ev}(t) - q_{\inf}(t), \tag{3}$$

where S_{cl} is the storage in the cover layer.

The infiltration flux is assumed to be a non-linear function of the storage in the cover layer.

$$q_{\inf} = -K_{cl}(S_{eff})^{b_{cl}} \tag{4}$$

where S_{eff} is the effective storage which ranges from zero to one and is defined as:

$$S_{eff} = \frac{S_{cl} - S_{cl_{\min}}}{S_{cl_{\max}} - S_{cl_{\min}}}$$

where K_{cl} is the hydraulic conductivity of the cover layer (m d^{-1}), $S_{cl_{\max}}$ is the maximum achievable storage in the cover layer, $S_{cl_{\min}}$ is the minimum storage in the cover layer above which water will still freely drain and b_{cl} is a dimensionless empirical parameter which is larger than 0. When b_{cl} is less than 1, drainage from the cover layer predominantly occurs at low effective storage values, whereas if it is larger than 1, drainage predominantly occurs at high effective storage values.

The actual evapo-transpiration is calculated from the potential evapotranspiration:

$$q_{ev} = E_{pot}\ \ C_f\ \ f_{red}, \tag{5}$$

where E_{pot} is the potential evaporation [m/day], C_f is an empirical crop factor which is assumed to be a landfill specific constant and f_{red} is a factor allowing evapo-transpiration to be reduced in order to prevent the storage in the cover layer to become negative.

This model assumes that flow is caused by gravity only, that is, gradients in the hydraulic head of the soil do not drive water flow. The magnitude of flow is strongly controlled by the storage in the cover layer.

2.1.2. Water Balance of the Waste Body

The water balance of the waste body is calculated as:

$$\frac{\mathrm{dS_{wb}(t)}}{\mathrm{d}t} = q_{\inf}(t) - q_{leach}(t), \tag{6}$$

where q_{leach} is the leachate flux from the waste body to the drainage system.

We can rewrite Equation 6 as a function of life expectancies using a probability distribution of life expectancies $p_{S_{wb}}(T_E, t)$ using:

$$\frac{\mathrm{d}S_{wb}(T_E,t)}{\mathrm{d}t} = \frac{\partial S_{wb}(T_E,t)}{\partial t} + \frac{\mathrm{dT_E}}{\mathrm{d}t}\frac{\partial S_{wb}(T_E,t)}{\partial T_E},$$

to get

$$\frac{\mathrm{dS_{wb}(T_E,t)}}{\mathrm{d}t} = q_{\inf}(t) p_{q_{\inf}}(T_E,t) - q_{leach}(t) \tag{7}$$

where

$$q_{leach}(t) = S_{wb}(0,t) \tag{8}$$

which states that the leachate flux is equal to all the water present in the waste body with a life expectancy of 0 at time t. Please note that after we have solved for $S_{wb}(T_E,t)$ we can calculate $p_{S_{wb}}(T_E,t)$ with:

$$p_{S_{wb}}(T_E,t) = \frac{S_{wb}(T_E,t)}{S_{wb}(t)}.$$

In order to numerically solve Equation 7, we need to discretize along life expectancies (T_E). T_E can range from 0 to infinity. To simplify implementation we chose to discretize T_E in a discrete number of daily values from $T_{E,0}$ to $T_{E,n_{tt}}$. $S_{wb}(T_{E,i},t)$ represent water storage cells with a life expectancy $T_{E,i}$ at time t. $S_{wb}(T_{E,n_{tt}},t)$ represents a bulk cell containing all water with a life expectancy larger than n_{tt} days. All water with $T_E > T_{E,n_{tt}}$ is assumed to be added to the storage in the bulk of the waste body S_{bulk}:

$$S_{bulk}(t) = S_{wb}(T_E \geq T_{E,n_{tt}},t) \tag{9}$$

The consequence of this choice is that an additional water flux needs to be added to the model the rate of change in $S_{bulk}(t)$. We call this flow, base flow and assume that it is a function of S_{bulk}. The base flow (q_{bF}) controls the leachate flow from the landfill in prolonged drought periods, however, it cannot continue indefinitely because of the finite amount of water stored in the bulk of the waste body. To take this finite amount in to account, we apply a gamma distribution function in order to allow q_{bF} to reduce gradually after the bulk storage reaches a critical level:

$$q_{bF}(S_{bulk}) = q_{bF_0}\frac{f_{bF}(S_{bulk})}{S_{bF}}, \tag{10}$$

where

$$f_{bF}(S_{bulk}) = \frac{x^{\sigma_{bF}-1}e^{-(S_{bulk}-S_{bulk,\min})}}{\Gamma(\sigma_{bF})}$$

where $S_{bulk,\min}$ is the minimal storage in the bulk where the base flow drops to zero, S_{bf} (m) is a scaling factor for the bulk storage and σ_{bF} (m) determines the shape of the base flow function.

Including the base flow in Equation 7 leads to:

$$\frac{\mathrm{dS_{wb}(T_E,t)}}{\mathrm{d}t} = q_{\inf}(t)p_{q_{\inf}}(T_E,t) + q_{bF}(S_{bulk}(t))\,p_{q_{bF}}(T_E,t) - S_{wb}(0,t) \tag{11}$$

where we have two distinct sources for the water traveling through the waste body: (a) water infiltrating from the cover layer, $q_{\inf}$ and (b) the base flow released from the bulk of the waste, q_{bF}.

The probability distribution $p_{q_{\inf}}(T_E,t)$ describes the life expectancies along a large number of trajectories in the waste body along which water is moving. Following Rosqvist and Destouni (2000), we assume that the ensemble of the life expectancies of all particles infiltrating from the cover layer can be described with a bimodal log-normal probability density function. For this paper we assume that this bimodal probability distribution function is time invariant.

$$p_{q_{\inf}}(T_E) = \beta p_{q_{\inf},fast}(T_E) + (1-\beta)p_{q_{\inf},slow}(T_E) \tag{12}$$

$$p_{q_{\inf},i}(T_E) = \frac{1}{T_E\sigma_i\sqrt{2\pi}}\exp\left(\frac{-(\ln(T_E)-\ln(\tau_i))^2}{2\sigma_i^2}\right) \tag{13}$$

In these, $p_{q_{\inf},fast}(T_E)$ and $p_{q_{\inf},slow}(T_E)$ are log-normal probability distribution functions for fractions of water experiencing fast flow and slow flow respectively (indicated with index i in Equation 13) where β is the fraction of water following the fast flow probability distribution function.

The probability distribution function for the life expectancies of the base flow is assumed to be a time invariant gamma distribution, in which Γ is the gamma function and a_{bF} is a value between 0 and 1:

$$p_{bF}(T_E) = \frac{x^{a_{bF}-1}e^{-x}}{\Gamma(a_{bF})} \tag{14}$$

where x is a normalized life expectancy of the water released from the bulk waste defined as:

$$x = T_E / T_{E,\mathrm{norm}} \tag{15}$$

As stated before, assumptions underlying this approach are that the flow in the waste body is considered to be a causal process and that both travel time distributions are assumed to be time invariant. This implies that the flow occurs along specific paths or streamlines and that the flow rates in these paths remain constant in time. Harman (2015) shows that it is relatively simple to relax the assumption of time independent pdfs, for example, by making T_E a function of the storage.

2.1.3. Water Balance of the Drainage Layer

In the landfills analyzed for this paper, leachate is actively pumped from the drainage system by an automatic system that maintains the water level between a minimum and a maximum value. As a consequence, water levels in the drainage system are nearly constant and we can assume that the flux from the drainage system is identical to the flux entering the drainage system:

$$q_{drain} = q_{leach} \tag{16}$$

2.2. Solute Mass Balance

In order to demonstrate the concept we chose to consider only conservative solutes for this paper. This implies that the mass flux is fully controlled by water flow. Because the probability distribution of life expectancies implicitly takes account of the dispersion that occurs as water flows through the system we neglect solute diffusion as a separate process because it is very slow. The solute flux can therefore be written as:

$$\boldsymbol{q}_M = \boldsymbol{q}_w \, c \tag{17}$$

The solute mass balance of the cover layer can be calculated as:

$$\frac{\mathrm{d}M_{cl}(t)}{\mathrm{d}t} = c_{rain}(t)q_{rf}(t) - c_{cl}(t)q_{\inf}(t). \tag{18}$$

The solute mass balance can be defined with:

$$\frac{\mathrm{d}M_{wb}(T_E,t)}{\mathrm{d}t} = c_{\inf}(t)q_{\inf}(t)p_{q_{\inf}}(T_E,t) + c_{bulk}(t)q_{bF}(S_{bulk}(t))\,p_{q_{bF}}(T_E,t) - M_{wb}(0,t) \tag{19}$$

where M_{cl} and M_{wb} are the total mass of conservative species present in the cover layer and waste body (kg/m^2), respectively. As we only consider old landfills which no longer accept waste and we assume that the concentration of conservative solutes in rain in the Netherlands is much lower than those found in landfill leachate we take influx of solutes with rainfall to be zero. In the waste body each water parcel with a specific life expectancy is associated with a specific mass ($M_{wb}(T_E,t)$ similar to the storage in the waste body. The concentration of conservative species for the cover layer is:

$$c_{cl}(t) = \frac{M_{cl}(t)}{S_{cl}(t)},$$

and the concentrations of the water parcels in the waste body are:

$$c_{wb}(T_E,t) = \frac{M_{wb}(T_E,t)}{S_{wb}(T_E,t)}.$$

2.3. Solution Algorithm for Cover Layer

Implementing the equations for the cover layer as a differential equation results in a model that requires quite a lot of time to solve. In order to have a fast model which allows for a large number of runs in a Monte Carlo simulation framework, we chose to simulate the flows for the cover layer with an algorithmic implementation based on the equations in paragraph 4.1.1. Rainfall data and potential evapotranspiration data are available as daily average fluxes. The implementation is shown in Algorithm 1 using a time step Δt of 1 day.

Algorithm 1. Implementation of the Water Balance Algorithm for the Cover Layer

1. Estimate infiltration flux with Equation 4 and estimate the new storage

$$S_{cl_{est_{n+1}}} = S_{cl_n} + \left(q_{rf_{n+1}} - q_{\inf_n} + q_{Ev_{n+1}}\right)\Delta t. \tag{20}$$

2. If the estimated amount of water in the cover layer is larger than the maximum available storage capacity, $S_{cl_{est_{n+1}}} > S_{cl_{\max}}$, we need to increase the amount of infiltration by short circuiting the flow to the waste body and and then recalculating $S_{cl_{est_{n_1}}}$ with the corrected $q'_{\inf_n}$

$$q'_{\inf_n} = q_{\inf_n} + (S_{cl_{\max}} - S_{cl_{est_{n+1}}})/\Delta t, \tag{21}$$

$$S_{cl_{est_{n+1}}} = S_{cl_n} + \left(q_{rf_{n+1}} - q'_{\inf_n} + q_{Ev_{n+1}}\right)\Delta t. \tag{22}$$

3. However if ($S_{cl_{est_{n+1}}} < 0$ and $S_{cl_n} > S_{cl_{\min}}$) then we need to limit the amount of infiltration (there is not enough water in the cover layer to sustain infiltration) and perhaps also reduce the amount of evaporation:

$$q'_{\inf_n} = (S_{cl_{\min}} - S_{cl_n})/\Delta t \tag{23}$$

after which we again calculate $S_{cl_{est_{n+1}}}$ using equation 22.

4. If the estimated storage after correction is still negative, ($S_{cl_{n+1}} < 0$), we need to limit the amount of evaporation and then recalculate $S_{cl_{est_{n+1}}}$

$$q'_{Ev_{n+1}} = q_{Ev_{n+1}} + S_{cl_{n+1}}/\Delta t, \tag{24}$$

$$S_{cl_{est_{n+1}}} = S_{cl_n} + \left(q_{rf_{n+1}} - q'_{\inf_n} + q'_{Ev_{n+1}}\right)\Delta t. \tag{25}$$

This approach implicitly implements the f_{red} term in Equation 5.

5. All conditions should now be fulfilled so we have a new update of the storage in the cover layer:

$$S_{cl_{n+1}} = S_{cl_{est_{n+1}}} \tag{26}$$

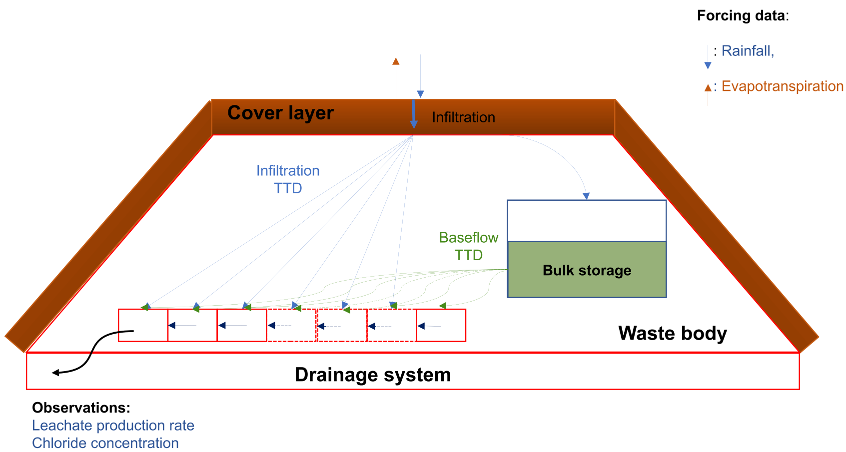

Figure 1. Illustration of the conceptual model of the landfill system. The cover layer allows rain water to be buffered so that evaporation can also occur on days without rainfall. Water subsequently infiltrates in to the waste body. In the waste body the infiltrated water is distributed over a discrete number of life-expectancy cells and the remainder is added to the bulk. On a daily basis water in the cell with a life expectancy (T_E) of zero is emptied in the drainage system from where it is immediately removed as leachate.

2.4. Solution Algorithm for the Waste Body

In order to solve Equation 11 we discretize it over both time dimensions where we use a daily time step $dt = 1$ day and for each time step, the life expectancy is distributed over $n_{tt} + 1$ values ranging from 0 to n_{tt}. This approach is illustrated in Figure 1. Each day, water infiltrating from the cover layer is distributed among the cells in the waste body using $p_{q_{\text{inf}}}(T_E)$ and water flowing from the bulk as base flow using $p_{bF}(T_E)$.

At $t = 0$, all cells are initialized with an initial amount of storage S_{ini}. Then every daily time step, all cells are shifted in life expectancy, that is, the cell with $T_E = 1$ becomes 0, the cell with $T_E = n_{tt}$ becomes $n_{tt} - 1$ days and the cell with $T_E = n_{tt} - 1$ is filled with water from the bulk waste depending on the base flow.

2.5. Model Calibration Using Bayesian Inference

Before we can use this model to simulate leachate production rate and quality we need to quantify the parameters. The parameters can be obtained by history matching of simulated leachate volumes and leachate concentrations to those obtained from measurements. We inferred the values of the parameters with the Multiple-try DREAM(ZS) package Laloy and Vrugt (2012), implemented in pyDREAM Shockley et al. (2018). DREAM (Vrugt, 2016) applies a Bayesian inference scheme to obtain the distribution of model parameters ($\boldsymbol{\theta}$) which optimally describe the measured data in a probabilistic framework. Bayesian inference,

$$p(\boldsymbol{\theta}|\hat{\mathbf{y}}) \propto p(\boldsymbol{\theta}) \cdot L(\boldsymbol{\theta}|\hat{\mathbf{y}}), \tag{27}$$

allows us to calculate the joint posterior probability distribution ($p(\boldsymbol{\theta}|\hat{\mathbf{y}})$) of the set of parameters using the measured data. The posterior distribution is calculated using the prior distribution of the parameters ($p(\boldsymbol{\theta})$) and the likelihood of the parameters given the measured data ($L(\boldsymbol{\theta}|\hat{\mathbf{y}})$).

For the likelihood function we applied the generalized likelihood function proposed by Schoups and Vrugt (2010):

$$L(\boldsymbol{\theta}|\hat{\mathbf{y}}) = -n \ln \frac{2\sigma_\xi \omega_\xi}{\xi + \xi^{-1}} - \sum_{t=1}^{n} \sigma_t - c_\beta \sum_{t=1}^{n} |a_{\xi,t}|^{2/(1+\beta)}. \tag{28}$$

For a detailed description and explanation of the parameters in this function we refer to Schoups and Vrugt (2010). We chose to use the generalized likelihood function because it allows for an improved handling of residual errors which can be correlated, heteroscedastic and non Gaussian with varying degrees of kurtosis and skewness. As a result, this approach allows for a correct statistical description of the data and residual errors, without the need for separating the different error sources. In Equation 28, we defined the measurement error as $\sigma_t = \sigma_0 + \sigma_1 y_t(\boldsymbol{\theta})$, $a_{\xi,t}$ is an independently and identically distributed random error with zero mean and unit standard deviation, described by a skew exponential power (SEP) density using parameters ξ and β to account for non-normality, scalars ω_β, σ_ξ, and c_β are derived from values of ξ and β which are a skewness and kurtosis parameter respectively and $\boldsymbol{\phi} = \{\phi_1, \ldots, \phi_4\}$ stores coefficients for an auto-regressive model of error residuals.

The generalized likelihood function is based on an additive non-linear regression model:

$$\mathbf{Y} = \mathbf{E} + \mathbf{e}$$

where $\mathbf{Y}$ is a vector of n observations, $\mathbf{E}$ is a corresponding vector of expected values; and $\mathbf{e}$ is a vector of zero mean random errors or residuals. The vector $\mathbf{e}$ includes measurement error, model input, model structural errors. In order to account for heteroscedastic errors, Schoups and Vrugt (2010) suggest to include multiplicative bias factors in order to account for systematic deviations in model predictions:

$$E_t = Y_{h,t}(\mathbf{X}|\boldsymbol{\theta}_h)\mu_t. \tag{29}$$

In this equation we assume that expected values can be modeled with a mass-balanced base flow model h, which yields simulated values $\boldsymbol{Y}_h$ as function of an observed input $\boldsymbol{X}$ and a vector of model parameters $\boldsymbol{\theta}_h$. In this equation the simulated flow $Y_{h,t}$, and bias factor μ_t vary as a function of time. Schoups and Vrugt (2010) suggest to amplify the non-linearity in the response of the leachate production using

$$\mu_t = \exp(\mu_1 Y_{h,t}), \tag{30}$$

but we found that we obtained the best results by not including this bias factor so we kept μ_1 to zero.

As suggested by Vrugt (2016), we include $\sigma_0, \sigma_1, \beta, \xi, \boldsymbol{\phi}$ and $y_{\min}$ as so-called nuisance variables in the inference together with all the other unknown parameters.

The generalized likelihood function was applied to both the times series of leachate production volumes and concentration data. The prediction of the cumulative total was constrained by adding a third normal likelihood term based on the cumulative leachate production over the inference period as a likelihood based on a sum of squares:

$$L_{cum}(\boldsymbol{\theta}|\hat{y}_{cum}) = -\frac{1}{2}\ln(y_{cum} - \hat{y}_{cum})^2. \tag{31}$$

The total liklihood, L_{tot} is the sum of the three likelihood values.

2.6. Boundary Conditions

The model is driven by daily rainfall and evaporation data which we downloaded from the Royal Dutch Meteorological Institute (KNMI, n.d.).

2.7. Initialization of the Model

Initial values for the model states need to be defined before we can keep track of the change in states which are driven by the varying boundary conditions. The important states are the storage in the cover layer, S_{cl}, the storage in the waste body, S_{wb} and the solute mass in the cover layer and waste body respectively, M_{cl} and M_{wb}. The model is started in the past well before measurements become available so that the effect of the initial conditions have been minimized by the seasonally varying boundary conditions. For the scenarios presented here, we start the simulation on the first of January 2003. The model is driven by daily precipitation and potential evapotranspiration data.

In order to facilitate a physical interpretation of the initial storage states we relate them to the average water filled porosity and heights of the cover layer and waste bodies. In addition we estimate the initial mass present in the cover layer and waste body using an average concentration. The effect of initializing with average values will become smaller over time due to the cyclic seasonality in the meteorological boundary conditions.

The initial states are calculated with:

$$S_{cl} = \theta_{w_{cl}} * H_{cl} \tag{32}$$

$$S_{wb} = \theta_{w_{wb}} * H_{wb} \tag{33}$$

$$M_{cl} = S_{cl}\ c_{cl} \tag{34}$$

$$M_{wb} = S_{wb}\ c_{wb} \tag{35}$$

where subscripts cl and wb indicate cover layer and waste body. $\theta_{w_{cl}}$ and $\theta_{w_{wb}}$ are the volumetric water contents in the cover layer and waste body [-], H_{cl} and H_{wb} are the thickness of the cover layer and waste body $[m]$. M_{cl} and M_{wb} are the solute masses in the cover layer and waste body per unit landfill area. The maximum saturation of the cover layer is then parameterized using the maximum volumetric water content ($\theta_{w_{cl},\max}$) which is equal to the porosity of the cover layer. In order to ensure that the minimum storage in the cover layer is always less the maximum storage, it is initialized as a fraction of $\theta_{w_{cl},\max}$: $f_{w_{cl},\min}$. The saturation of the cover layer is initialized as to be half the difference between $S_{cl_{\max}}$ and $S_{cl_{\min}}$, and the saturation of the waste body was initialized directly. The minimum bulk storage, $S_{bulk_{res}}$ is parameterized in a similar manner with $f_{w_{wb},\min}$.

Each life expectancy cell and the bulk storage in the waste body are initialized with the same initial concentration from which the initial mass present in the waste body is calculated. The amount of mass removed every time step with the leachate is the amount of mass present in the cell with a life expectancy of 0 days. Mass can only enter the life expectancy cells from the bulk with the base flow. The mass in the bulk is updated every time step with the amount removed with the base flow. The mass in the life expectancy cells remains constant with time, infiltrating water from the cover layer only leads to dilution of the concentration.

3. Site Specific Data and Prior Distributions of Unknown Parameters

3.1. Landfills

All data are from two landfills which are currently part of the Natural Biodegradation Research Program on Dutch Landfills (Stichting Duurzaam Storten, 2017). The Wieringermeer landfill is near Medemblik and the Braambergen landfill is near Almere, both in the Netherlands. Details for these landfills are given in Table 1. Both landfills are operated by Afvalzorg N.V.

Geo-referencing available as-built drawings to background maps and recent high resolution areal photographs in a GIS package, allowed us to estimate the surface area of the basal drainage system and the heights and surface areas of the landfills. The heights of the basal drains have been measured in 2016. As the topography of the sites is variable, we estimated the volume of the waste body from the GIS derived data and then calculated the average height of the waste body by dividing the volume by the area of the basal drainage system. Background data for the four landfill cells can be found in Table 1.

Detailed monitoring of produced leachate volumes and leachate quality is carried out in the context of the biodegradation research program by the landfill operator since March 2012.

Data available for model calibration and verification were the cumulative leachate production measured every 15 min from 14 June 2012–1 November 2024 and chloride concentrations measured in a commercial laboratory over the same period. Laboratory analyses were performed on leachate samples that were taken once every 2 weeks.

By testing different attempts to infer the parameters from the data, we found that the best results were obtained when cumulative leachate production is transformed to weekly leachate production rates by differentiation of the cumulative production. This transformation allows the model to capture the weekly dynamics in the data.

Rainfall and potential evaporation data are downloaded from automatic weather stations operated by the Royal Dutch Meteorological Institute (KNMI, n.d.). For the Wieringermeer landfill we used the data from the Berkhout

Table 1
General Background Information on the Landfill Cells

Town	Braambergen Almere	Wieringermeer Medemblik
Compartment ID	11N, 11Z and 12	VP06
Area basal drainage system (m^2)	**11N:** 34,802 **11Z:** 35,188 **12:** 30,000	28,355
Time in operation	1,999–2008	1,992–1,998
Total amount of waste [ton]	1,216,723	281,083
Volume of landfill cell (m^3)	**11N:** 345,426.2 **11Z:** 388,981.9 **12:** 315,254.4	323,094
Average Height of the landfill (m)	**11N:** 9.9 **11Z:** 11.1 **12:** 10.5	12.2
Current landfill cover	Soil, incinerator bottom ash, jet grout (1.5 m)	Soil (1.5 m)
Landfill gas extraction	36 gas wells	2 gas wells + 1 shared
Bottom liner	Combination of mineral (50 cm sand bentonite) and 2 mm HDPE foil	Single 2 mm HDPE foil
Leachate drainage and collection	3 separate drainage systems for **11N**, **11Z** and **12** (3 pump pits)	Separate drainage system for compartment 6, single pump pit
Aeration system	Installed between Nov 2016-Jan 2017 and expanded in 2022 **11N**: well spacing: ca. 20 m, after expansion about 10 m (total number: **11N**: 65, after expansion 113) **11Z** well spacing: ca. 15m after expansion about 7 m (total number: 114 after expansion 163) **12** well spacing: ca. 20m (total number: 56) filter length: 2m depth filter: 1 m above the top of the drainage system	Installed in Oct 2016 well spacing: ca 14 m total number gas wells: 109 filter length: 2 m depth filter: 1.4 m above the top of the drainage system

station, for Braambergen we used the data from the Lelystad station. Daily rainfall, and calculated reference evapo-transpiration were used for the water balance analysis.

Between October 2016 and February 2017 a large number of wells were installed at both landfills through which the waste body is aerated. As the aim of aeration is to ultimately improve leachate quality, the filters of the wells are installed deep in the waste body (about 1–2 m above the top of the drainage system. In 2023 additional wells were installed at Braambergen.

3.2. Prior Distribution Ranges

In order to use Bayesian inference we require prior distributions of uncertain model parameters. We chose to initialize the optimization with uniformly distributed priors over a predefined search range (Table 2). The initial ranges were defined based on the expected physical values and sometimes by trial and error. In this last case, when it became apparent during the optimization that the initial prior was too constrained, the boundaries of the distribution were extended. For priors where variation is expected to vary across orders of magnitude, the ranges were initialized using the log-10 values of the parameters.

The number of finite travel times (n_{tt}) was set to be equal 1,825 days (or 5 years). The model bias factor μ_1 in Equations 29 and 30 was set to zero as first inference attempts indicated that model bias was not important.

In order to minimize the effect of the initial parameters on the calibration with measured data we started the model on 1 January 2003. This 'burn-in' time, allows for the base flow from the bulk that initiated on 1 January 2003, to

Table 2
Priors for the Parameter Distributions Used in the Bayesian Inference

Parameter	Minimum	Maximum	^{10}log
Cover layer			
C_f	0.75	1.5	–
$\theta_{w_{cl},\max}$	0.3	0.5	–
$f_{w_{cl},\min}$	0.0001	1	–
K_{cl}	−5	3	x
b_{cl}	0	8	–
$c_{ini_{cl}}$	−4	−4	x
Waste body			
τ_{fast}	1	730	–
σ_{fast}	−5	2.5	x
$\Delta\tau_{slow}$	0	9,125	–
σ_{slow}	−5	3	x
β_f	0	1	–
$\theta_{wb_{ini}}$	0.0	0.5	–
$f_{wb_{\min}}$	0.0	0.5	–
bF_0	−5	−2	x
$c_{ini_{wb}}$	2	6	x
S_{bF}	0	15	–
σ_{bF}	0	15	–
a_{bF}	−9	0	x
t_{norm}	0	1,825	–
Generalized likelihood			
σ_0	−8	2	x
σ_1	−8	2	x
μ_1	0		–
β	−1	1	–
ξ	0.1	10	–
$\phi_{1,leachate}$	−1	1	–
$\phi_{1,conc}$	−1	1	–
$[\phi_2, \phi_3, \phi_4]$	[0, 0, 0]		–

Note. Priors are uniform distributions. The column^{10} log indicates which parameters have a range defined by the^{10} log values. Parameters with only a minimum value have not been inferred, the given values are used as a constant. The parameters for the generalized likelihood function are the same for both the leachate flow and the concentration data, except for the correlation parameter ϕ_1 which were maximized based on trial and error.

have moved completely across all 1,825 cells before the results are compared to any measurement values. As such the solute mass and volume of water in the travel time cells are by then fully constrained by the base flow from the bulk waste 5 years earlier.

The model was calibrated using measured leachate volumes and chloride concentrations from the first of January 2014 to the 31st of December 2020. The remaining available data from the complete data set from 12 June 2012 until 1 November 2024 were used to assess the model performance beyond the calibration range.

Calibration was initially started allowing the pyDREAM to randomly sample from the prior distributions. Once the inference had converged, pyDREAM was restarted with the final values of the 3 chains as an initial guess until the Gelman Rubin criterion indicating convergence was met (Vrugt (2016)). The data were analyzed using the final distributions from this last optimization. The convergence and consequently the parameter distributions were assessed using the final 50% of the length of each chains Vrugt (2016). The optimal parameter set is the Pareto optimal of the three likelihoods calculated for leachate cumulative outflow, leachate outflow rate and leachate concentration.

4. Results and Discussion

4.1. Simulated Leachate Outflow Rate and Leachate Concentration Values

After convergence in pyDREAM we may assume that all likelihood values in the final parameter set are samples from the distribution of likelihood values in the model that fit the measurements best. Because the inference is carried out in a hyperspace with 29 dimensions, different parameters can be highly correlated with each other. This is illustrated in Figure S1 in Supporting Information S1 for some of the parameters in the converged set for the Braambergen 11N waste body that have a Pearson's correlation coefficient larger than 0.75. The presence of a strong correlation between parameters is an indication that the range of some parameters can cover the complete prior-range. The parameter inference procedure has sampled the correlation in the posterior joint probability distribution. As a result, we cannot use averaged parameter values from the posterior joint probability distributions. Instead, we need to select individual parameter vectors from the total converged distribution of parameters.

In order to provide insight in how well the model is able to describe the measured data we decided to present model estimations based on the posterior likelihood for parameters vectors where the density is 1, and close to 0.95, 0.5, and 0.05 from the converged distribution. The cumulative distribution of likelihood values shown Figure S2 (Supporting Information S1) for the four waste bodies analyzed for this paper. The likelihood values corresponding to the probability quantiles of the lines in the following graphs are reported in the legends of the figures.

The results of the simulated values of leachate production and leachate chloride concentration are compared with measured values for the Braambergen 11N waste body in Figure 2. In the Supporting Information (Figure S3 in Supporting Information S1) we present similar figures for the Braambergen 11Z and 12 and the Wieringermeer VP06 waste bodies.

The generalized likelihood model of Schoups and Vrugt (2010) allows us to calculate a simulated measurement error with the forward model. For all sites the model is able to describe cumulative leachate production and

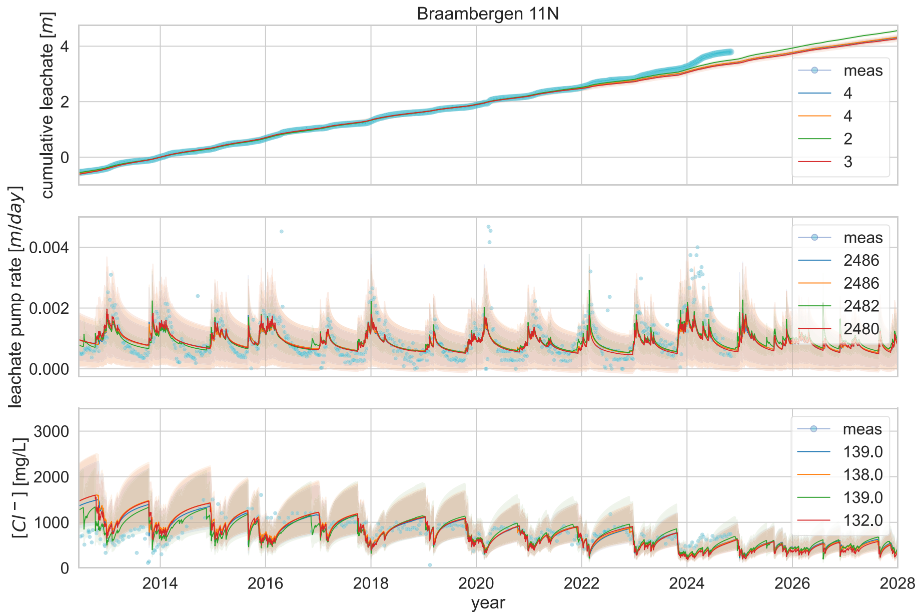

Figure 2. Simulated and measured values of cumulative leachate production (CL [m]), leachate pump rate (LPR [m/day]) and leachate chloride concentration (conc $\left[\mathrm{kg/m^3}\right]$ for Braambergen 11N. The four colors are the results of four scenarios corresponding to distinct values of likelihood from the converged parameter set. The shaded areas are the 95% confidence intervals estimated from the generalized likelihood model. The legends give the likelihood of the parameter set for the three objective functions used for optimization.

leachate flow dynamics with similar accuracy. The uncertainty estimated with the 95% confidence intervals is able to capture the spread in the measurement data for the data set used for the calibration.

In the generalized likelihood model of Schoups and Vrugt (2010), heteroscedasticity is explicitly accounted for by assuming that the measurement error increases linearly with the expected value:

$$\sigma_t = \sigma_0 + \sigma_1 E_t.$$

The inferred parameters in the generalized likelihood model ($\sigma_{0,leachate}$, $\sigma_{1,leachate}$, $\sigma_{0,conc}$, and $\sigma_{1,conc}$) of which the log10 values are reported in Table 3, allow us to calculate the estimated measurement error for the simulated leachate production rates and leachate concentrations. For a leachate production rate of 1×10^{-3} m/day the measurement error parameter set with the highest likelihood will be 0.72×10^{-3} m/day or about 72% of the expected value. This implies that the approximate 95% confidence interval ranges between -0.41×10^{-3} and 2.41×10^{-3} m/day. The negative value is a consequence of the simplified approach for estimating the confidence interval.

For the concentration data this calculation is less straight forward because the measurement error is estimated using the log10 transform of the measured concentration data. The measurement error for the log10 of the concentration for the highest likelihood with an expected value of 1,000 mg/L is 0.11, or about 4% of the log10 transformed concentration. This leads to a 95% confidence interval between 612 and 1,634 mg/L. Similar values can be estimated for the other waste bodies as is graphically shown in Figure S3 (Supporting Information S1).

Although the uncertainty in the simulated values is significant, the estimates of the expected values give a good description of both measured time series. Please note that the calibration was carried out using data that were measured between 1-1-2014 and 31-12-2020.

The estimates of the leachate production rates do not capture the extremes in the measured data. This is due to several reasons. The first is because the optimal parameter set is a pareto optimum of both leachate production rates and chloride concentrations. The simulation represented by the red line in Figure S3 (Supporting Information S1) for the Braambergen 11Z waste body is a clear example, with a high likelihood for the concentration

Table 3
Statistics and Optimal Parameter Values After Optimization With DREAM(ZS), Braambergen 11N, Number of Samples in Converged Parameter Set = 60,000

	Mean	Std	Min	25.00%	50.00%	75.00%	Max	*1*	0.975	0.5	0.025
C_f	0.98	0.08	0.80	0.93	0.98	1.04	1.17	1.06	1.03	0.88	0.98
$\theta_{w_{cl,\max}}$	0.31	0.01	0.30	0.31	0.31	0.32	0.38	0.32	0.31	0.32	0.31
$f_{w_{cl},\min}$	0.19	0.03	0.11	0.17	0.20	0.22	0.26	0.20	0.22	0.23	0.25
$^{10}\log K_{cl}$	1.76	0.66	−0.53	1.26	1.76	2.28	2.99	2.15	1.20	2.17	−0.08
b_{cl}	3.48	1.35	0.85	2.67	3.21	3.80	8.00	2.90	2.00	3.46	1.19
τ_{fast}	297.04	191.65	26.05	135.26	241.12	453.54	727.53	244.97	171.36	162.45	140.90
$^{10}\log \sigma_{fast}$	0.44	0.09	0.18	0.36	0.44	0.51	0.69	0.40	0.36	0.41	0.39
$\Delta\tau_{slow}$	5092.93	2058.14	1007.65	3182.48	5063.49	6673.43	9112.41	7063.72	6855.75	1933.23	2796.92
$^{10}\log \sigma_{slow}$	−1.86	1.57	−4.97	−3.10	−1.94	−0.55	0.94	−1.07	−2.71	−3.01	−4.33
β_f	0.71	0.14	0.33	0.61	0.70	0.81	0.99	0.78	0.68	0.73	0.71
$\theta_{wb_{ini}}$	0.45	0.03	0.36	0.43	0.45	0.47	0.50	0.46	0.43	0.42	0.43
$S_{\min}$	0.38	0.07	0.17	0.33	0.38	0.44	0.50	0.38	0.35	0.45	0.40
$^{10}\log bF_0$	−1.90	0.23	−2.73	−2.03	−1.86	−1.73	−1.50	−1.89	−1.65	−1.86	−2.09
$^{10}\log c_{ini_{cl}}$	−0.49	2.26	−4.00	−2.39	−0.74	1.00	4.87	−3.48	−2.91	0.31	−3.70
$^{10}\log c_{ini_{wb}}$	3.39	0.05	3.30	3.35	3.38	3.41	3.60	3.38	3.41	3.41	3.40
S_{bF}	7.47	4.28	0.28	3.12	8.25	11.11	14.99	10.14	14.19	9.80	9.69
σ_{bF}	1.80	0.48	1.11	1.47	1.69	1.95	3.34	1.45	1.45	1.67	1.15
$^{10}\log a_{bF}$	−4.76	2.39	−8.99	−6.80	−5.05	−2.69	−0.50	−5.77	−4.50	−2.50	−7.16
$T_{E,\text{norm}}$	1265.17	391.84	5.84	1024.45	1333.88	1566.78	1824.89	1010.26	1122.62	1345.83	1500.61
$^{10}\log \sigma_{0,leachate}$	−3.63	0.04	−3.78	−3.65	−3.63	−3.61	−3.55	−3.63	−3.63	−3.69	−3.64
$^{10}\log \sigma_{1,leachate}$	−0.35	0.13	−0.75	−0.43	−0.35	−0.26	0.05	−0.43	−0.36	−0.63	−0.38
$\beta_{leachate}$	0.94	0.05	0.71	0.90	0.95	0.98	1.00	0.99	0.98	0.97	0.74
$\xi_{leachate}$	1.24	0.05	1.08	1.20	1.23	1.27	1.40	1.25	1.22	1.22	1.28
$\phi_{1,leachate}$	0.66	0.03	0.55	0.64	0.67	0.69	0.70	0.67	0.65	0.68	0.61
$^{10}\log \sigma_{0,conc}$	−1.03	0.05	−1.16	−1.06	−1.02	−0.99	−0.91	−1.01	−1.00	−0.96	−1.06
$^{10}\log \sigma_{1,conc}$	−5.16	1.68	−8.00	−6.63	−5.07	−3.68	−1.95	−5.45	−6.26	−4.43	−2.46
β_{conc}	0.93	0.05	0.75	0.91	0.95	0.97	1.00	0.97	0.97	0.95	0.78
ξ_{conc}	0.93	0.09	0.69	0.87	0.92	0.99	1.22	0.90	1.03	1.11	1.01
$\phi_{1,conc}$	0.59	0.07	0.36	0.54	0.59	0.64	0.70	0.59	0.53	0.67	0.58
L_{tot}	2569.35	4.88	2536.62	2566.29	2569.90	2572.43	2580.20	2580.20	2576.75	2569.90	2561.41

data, but a lower one for the leachate data. An important reason for this ambiguity lies in the fact that the model is an initial boundary value problem in which many of the waste body properties cannot vary with time. Early attempts, where first only leachate production data were used in the objective criterion, gave better fits of measured leachate production values, however attempts to subsequently describe the concentration data with fixed optimal parameters for the leachate production rate simulation led to poor results for the concentration data (results not shown). This implies that the assumption of purely convective flow for chloride is not completely true or that the parameters in the model vary with time. When inferring the parameters from both time series, leachate production rates need to be smoothed in the model to a certain extent. Another example of the initial boundary condition problem can be seen in the early predictions of the concentration data (before the start of the calibration period in 2014) where the simulated concentrations poorly describe the measured values for Braambergen 11N, 12 and Wieringermeer waste bodies.

The second reason is related to the quality of the measurement data. Leachate levels in the pump-pits of the waste body cells are allowed to vary over a narrow bandwidth of 10 cm. When the maximum level is reached, the pump will be switched on, when the minimum level is reached it switches off. During pumping, flow is cumulatively recorded. Since, the start of monitoring in April 2012, several problems were encountered with the pumps causing them to be switched off leading to a zero flow. As a consequence, the drainage system buffered the leachate. As soon the issues were fixed, the pumps quickly pumped the excess water until the set-points were reached. Pumping excess water led to temporarily much higher flow rates. In order to minimize this effect in the data, flow rates were obtained by taking backward differences of the cumulative data over a period of 7 days.

Because measurements of $q_{rf}(t)$ and $q_{leach}(t)$ were used to calibrate the model, the uncertainty in the water balance is mainly associated with the estimation of $q_{ev}(t)$. In the approach we used, the final estimate of $q_{ev}(t)$ is determined by multiple (correlated) parameters in the model. For most of the time $q_{ev}(t)$ is equal to the estimated potential evapotranspiration, but there are moments when evaporation became zero due to insufficient water present in the cover layer.

The posterior distributions of the model parameters indicate the model sensitivity of the system to the parameter values within the optimal likelihood parameter set. Figure 3 gives examples of posterior cumulative parameter distributions (in black) compared to the prior distributions in blue: the left column shows the inferred distributions for C_f, b_{F0}, and $c_{ini_{wb}}$ which are examples that have narrow distributions compared with the prior ranges given in Table 2. This narrow distribution implies that the measured data contains sufficient information to infer these parameters. The total water balance depends strongly on the crop factor, C_f, because the surface area of the waste body is fixed and the crop factor is the only parameter that can limit or enhance the evapotranspiration. Outflow concentrations strongly depend on the amount of solute mass present in the waste body, which is controlled by the initial concentration $c_{ini_{wb}}$ and the base flow, b_{F0}, which constrains the lowest flow rate during dry periods.

The parameters in the right column of Figure 3 are examples of poorly inferred parameters because the posterior distribution is very similar to the uniform prior distribution in Table 2. The outflow concentration does not depend on the initial concentration in the cover layer, $c_{ini_{cl}}$, in 2003. The simulation results show that any conservative solute in the cover layer is quickly removed. The posterior distribution can also be similar to the prior when the parameter is highly correlated with another one, a good example of this is the distribution of the mobile fraction in the cover layer, β_f which is highly correlated with τ_{fast} as shown in Figure S1 (Supporting Information S1). The last example of a poorly inferred parameter is the posterior distribution for $\Delta\tau_{slow}$ which does have a narrower range than the prior, however the final parameter range is still uniform over a wide range. The reason for this is that the inferred values cause the τ_{slow} value in Equation 13 to be larger than $n_{tt} = 1825\ days$. Given the way the model is implemented, any value larger than n_{tt} would lead to the same result.

4.2. Error Model

In order to check if the parameters in the error model are correctly inferred we follow the approach used by Schoups and Vrugt (2010) were different aspects of the residuals are evaluated. Figure 3 presents the results of this analysis for the Braambergen 11N waste body. For the error model we assume that the errors are heteroscedastic, the left plots show the residuals as a function of the expected values of measured leachate production rates and concentrations. The errors are nicely distributed around zero indicating that there is no bias in the results. The middle plots show that the distributions of errors are well described using the inferred parameters in the Skewed Exponential Power distribution of the generalized likelihood model. Clearly the error model does not follow a normal distribution. Finally, the autocorrelation in the residuals is adequately captured with the autocorrelation parameter even though the range for this parameter was constrained by a maximum value of 0.7. In addition, the different chains show similar results demonstrating that the final parameter set is indeed from a converged distribution (see Figure 4).

4.3. Extrapolation to 2066

The model allows us to make predictions of how emissions vary in the future while taking the uncertainty in the inferred model parameters in to account. The results for the Braambergen 11N waste body are shown in Figure 5, for the other waste bodies we refer to the Supporting Information S1, Figure S4. For all waste bodies, simulated results for selected parameter sets lie close to each other in the time range used to infer the parameters, but before and after the measurement time range, the results can diverge. This most clearly seen in the plot of the cumulative leachate production because small differences in simulated cumulate over time. For the time range used for the parameter inference these small differences are compensated, so the results for the different parameter sets do not

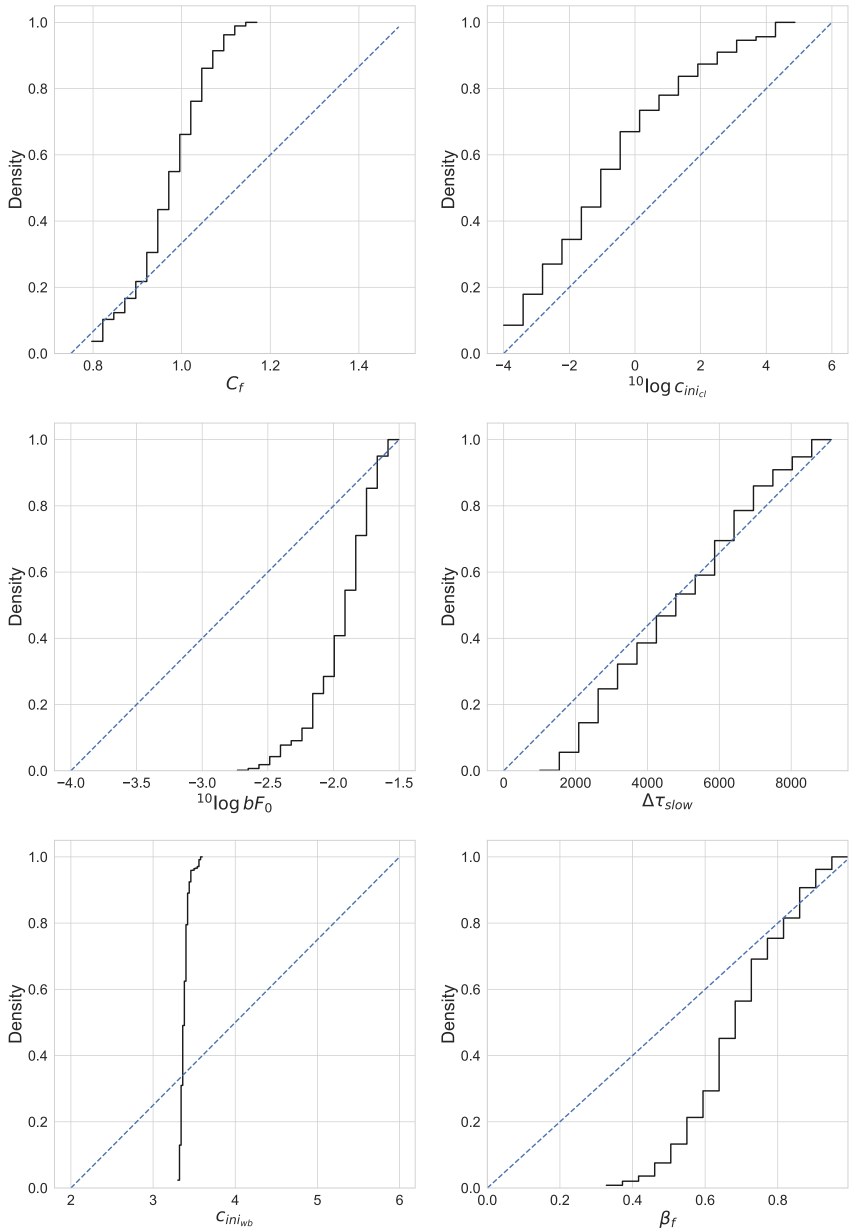

Figure 3. Posterior (black) and prior (dashed blue) cumulative distributions of selected parameters for Braambergen 11N. The left column gives examples of sensitive parameters that have converged to narrow posterior distributions, the right column gives examples parameters which are highly correlated with other parameters or which are not sensitive and therefore are close to the prior.

diverge from each other. Similar effects also occur for leachate production rates and leachate concentration data. However, as the effects do not cumulate as quickly as for the cumulative leachate production, it will take a much longer time series to see the effects in the graphs.

The simulated cumulative leachate production starts to deviate significantly from the measured cumulative flow after 2022 and in spring 2024 we see that measured flow rates fall above the 95% intervals. This is due to the fact the past years have moved from exceptionally dry to exceptionally wet in a very short time. An additional factor is that in 2022 additional wells were installed on Braambergen.

One of the most important assumptions in developing this approach is that the probability density functions of the travel time distributions do not change with time. It is likely that this assumption is too strict. Apparently, the

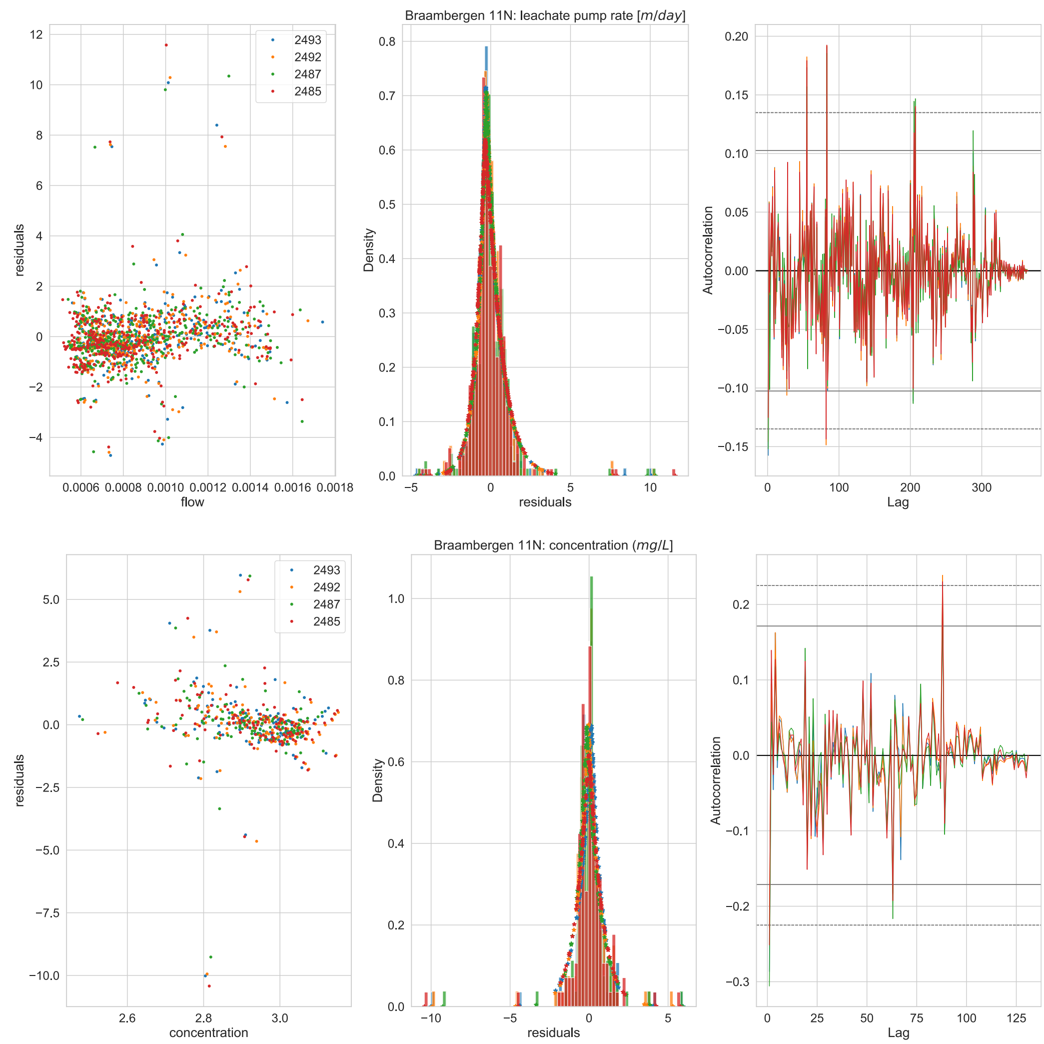

Figure 4. Braambergen 11 N. Plots of the residuals as a function of leachate production rate or concentration, distribution of the errors and auto-correlation in the errors for the flow data (top row) and concentration data (bottom row).

waste body is not able to store more water as rainfall increases, instead preferential flow to the drainage system seems to increase as well. Measurement artifacts also play a role. Because of the excess amount of water due to heavy rainfall, the landfill operator had to reduce the discharge to the water treatment plant by reducing the pump rate from the drainage system for a couple of weeks. The reduced flow of water may have led to a higher leaching of chloride.

In order to capture time dependent changes in the properties of the waste body and boundary conditions other approaches are required than used for this paper. Parameter distributions can be adjusted so that they become time dependent. This could be done by making them depend on the storage in the waste body as suggested by Harman (2015) or by inferring parameters and states in time using a particle filter as suggested by (Wang & Heimovaara, 2023).

The long-term predictions (Figure 5) indicate that chloride is gradually leached from the waste body and the concentration varies with the seasonal variation in infiltration. All scenarios (see also Figure S4 in Supporting Information S1) indicate that water storage in all waste bodies reach a long-term dynamic steady state where base-flow is compensated by storage from infiltration with life-time expectancies above n_{tt} or 1,825 days.

The concentration in the leachate is controlled by chloride mass and amount of water draining from the bulk volume of the waste body as base flow. The base flow rate is controlled by the volume of water stored in the bulk

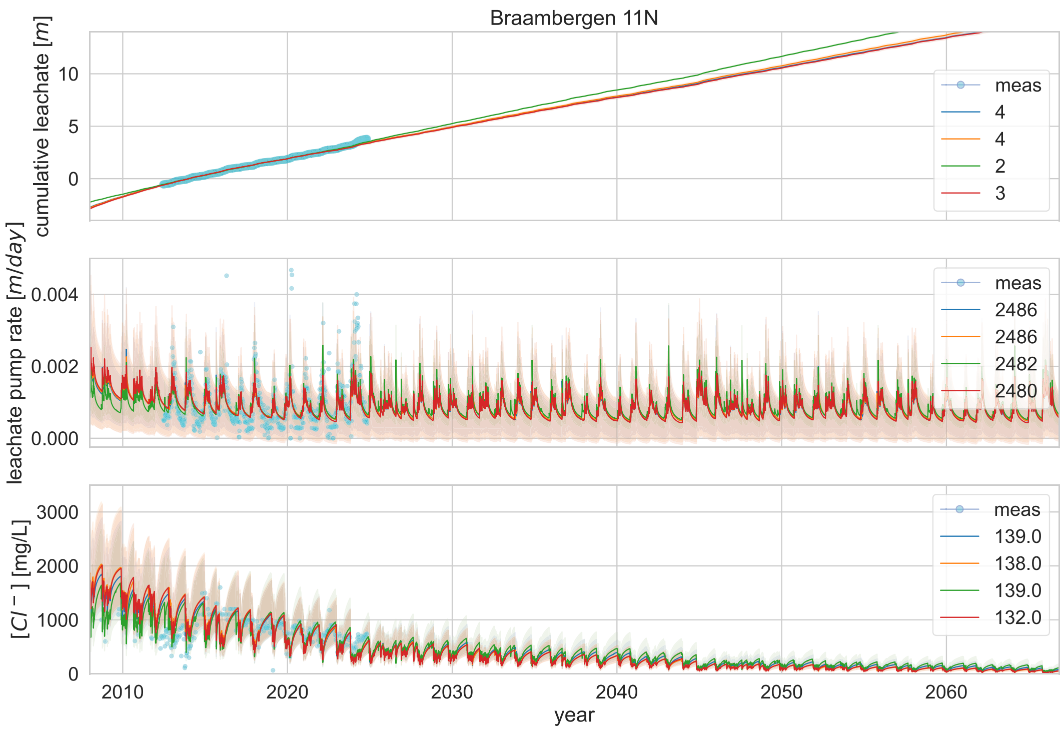

Figure 5. Long term extrapolation of cumulative leachate production (CL [m]), leachate pump rate (LPR [m/day]) and leachate chloride concentration (conc $\left[\mathrm{kg/m^3}\right]$ for Braambergen 11N. The scenarios are identical to the ones presented in Figure 2.

volume. Figure 6 shows the base flow as function of the bulk storage, Figure 7 the simulated base flow over time and Figure 8 shows the bulk concentration which is the ratio of the solute mass and storage. Each parameter set results in base flows with a different magnitude, however as the model is simulating both the water balance and the chloride mass-balance, final outcomes are very similar (see Figures 2 and 5). The differences in magnitudes for the base flow are caused by the fact that concentration is the ratio of solute mass over water storage. A higher concentration leads to a higher water mass, which therefore will lead to higher bulk storage values and therefore a different base flow function to achieve similar base flow mass rates. A clear indication of the mass balance are the similar magnitudes of the variations in the bulk storage (S_{bulk}) in Figure 8.

The oscillations in the base flow (Figure 6) can be explained by the seasonal variation in the infiltration leading to a variation in the bulk storage (Figure 8) and the slope of the base flow function (Figure 6).

4.4. Life Expectancy Distributions

In Figure 9, cumulative density functions are plotted for the life expectancy time distributions used to partition the water from the infiltration flux from the cover layer and the base flow from the bulk. Until a life expectancy of 2,500 days, the probability density functions for q_{inf} are completely determined by the fast flow fraction for all scenarios except the green one. Apparently the slow fraction only has an impact on water with a life expectancy which is much larger than 2,500 days. Given the fact that all water with a life expectancy older than $n_{tt} = 1825$ days is added to the bulk, the results imply that about 40% of the infiltrating water is added to the bulk of the waste body and that about 20% of the infiltrating water will have left the waste body as leachate after about 45 days. The same is true for the green scenario as the jump occurs after 1,900 days.

The cumulative density function for the base flow indicates that in all scenarios, except for the 50% probability scenario (green with a likelihood of 2487) are directly added to the cell with a life expectancy of 1,825 days which is consistent with convective transport with the water.

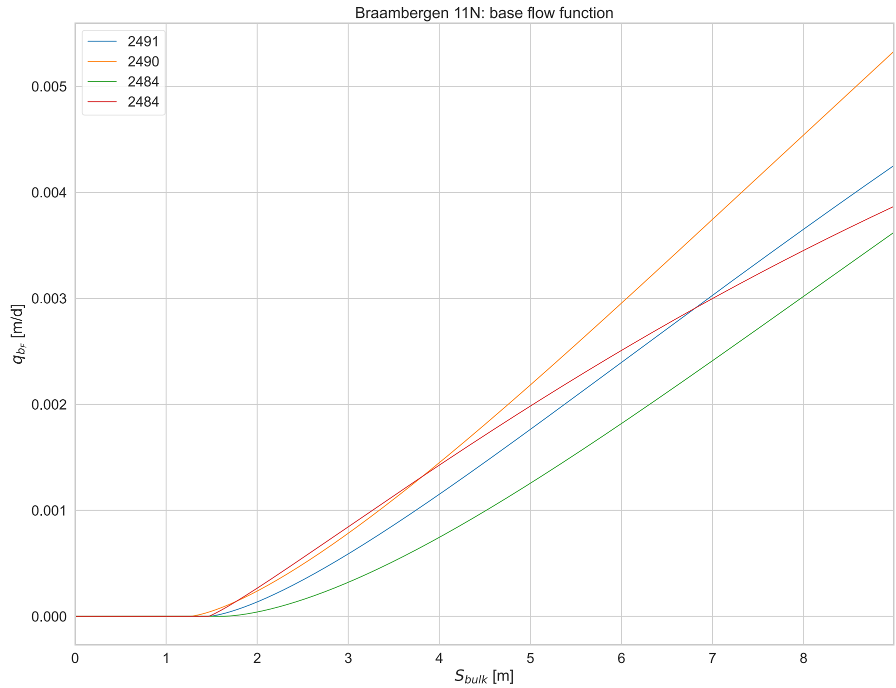

Figure 6. Base flow (b_F) functions for Braambergen 11N using Equation 10.

Because 20% of the water infiltrating from the cover layer has a life expectancy of less than 45 days, leachate concentration dynamics are dominated by this water moving preferentially through the waste body. This preferential flow explains why in winter concentrations are low and in summer concentrations are high. In the Netherlands, evapo-transpiration is high in summer leading to no or very low infiltration fluxes from the cover layer and solute present in cells with short life expectancy is not diluted. In winter evapo-transpiration is very low, leading to high infiltration fluxes which significantly dilute the solute present in the cells with short life expectancy.

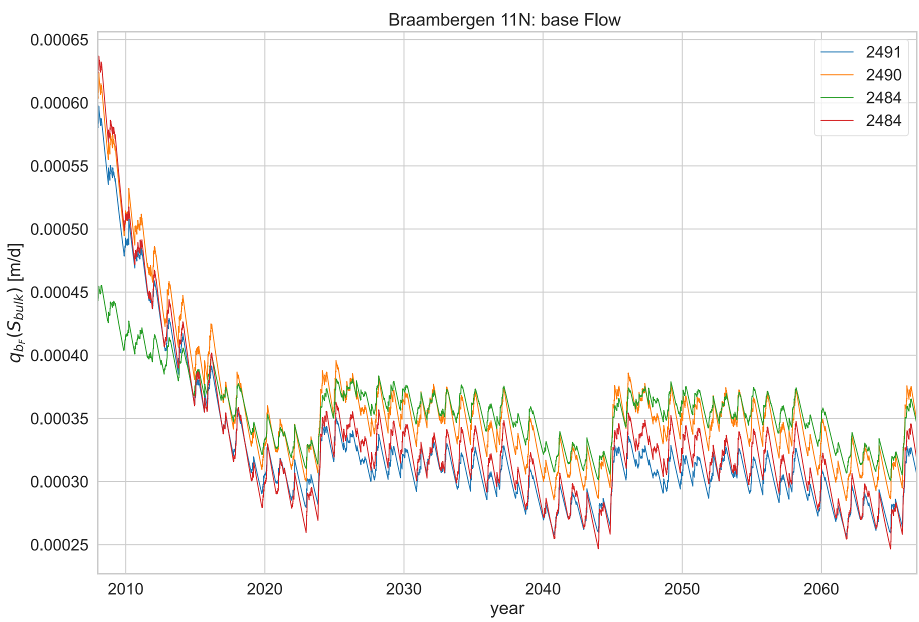

Figure 7. Simulated base flow (b_F) for Braambergen 11N.

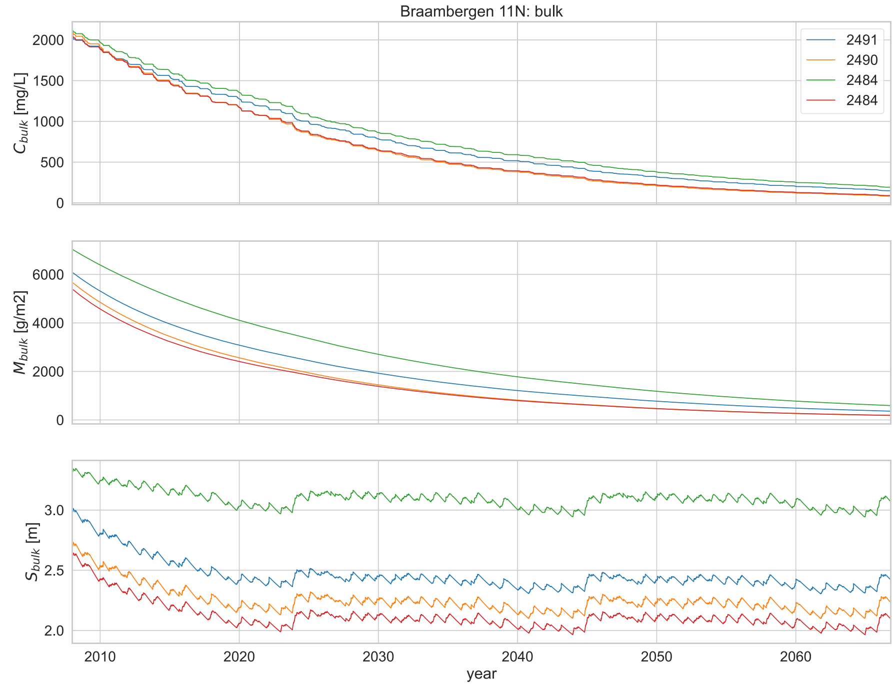

Figure 8. Simulated chloride concentration, mass and total storage in the bulk of the waste body for Braambergen 11N.

4.5. Emission Potential

The model keeps track of the mass balances of water and chloride in the waste body. The dynamics in the long term leachate production rates and leachate chloride concentrations are controlled by the mass present in the bulk. The only source for chloride in the leachate is the mass present at initialization, water is added via precipitation and subsequent infiltration in to the waste body. These assumptions lead to a gradual decrease in chloride mass which then leads to a gradual reduction in leachate concentrations as well.

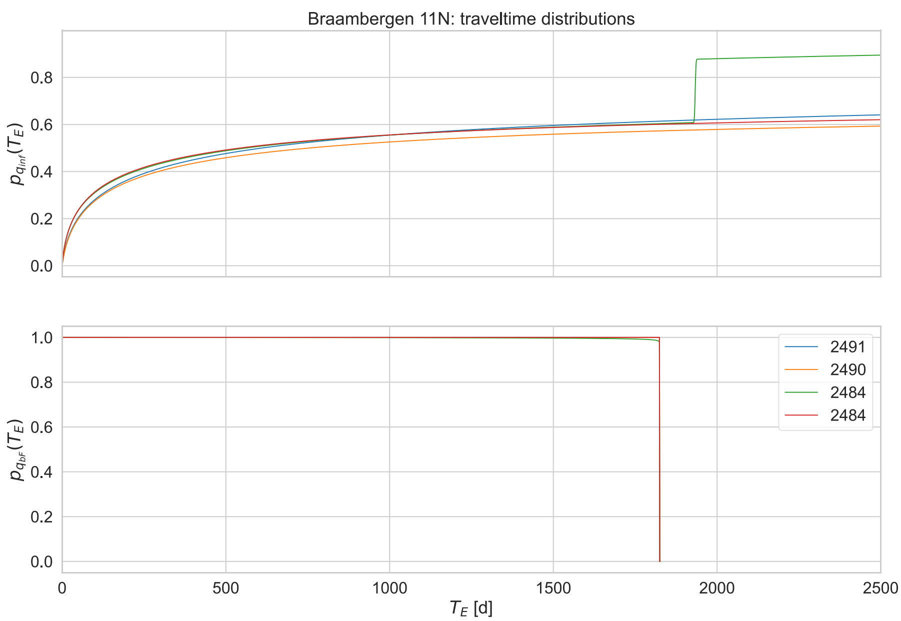

Figure 9. Cumulative densities of the travel time distributions for the infiltration flux from the cover layer (q_{inf}) using Equation 12 and the base flow from the bulk (q_{bF}) using Equation 14.

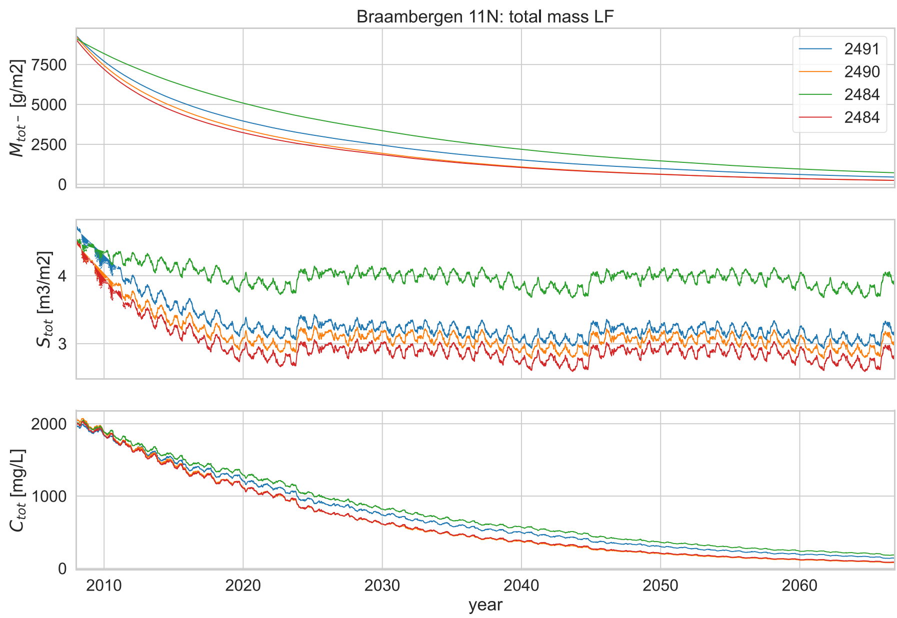

Figure 10. Total chloride mass, total storage and solute concentration in the waste body of Braambergen 11N.

Figure 10 shows the simulated totals of chloride mass, water storage and corresponding chloride concentrations for the waste body of Braambergen 11N. Clearly the mass shows an exponential decrease with time, which is to be expected. For the Braambergen 11N case, the difference in estimated total mass, varies from about 9,200 ± 100 g/m^2 in 2008 to 510 ± 250 g/m^2 in 2066. The corresponding emission from 2008 to 2066 ranges from 8,364 tot 9,018 g/m^2.

The leachate volume in the waste body decreases from 4.5 m to values between 2.6 and 4 m. The concentration is the ratio of the mass and the storage. As long as base flow occurs, the total mass in the waste body will decrease. The rate of decrease is controlled by the magnitude of the base flow and the concentration in the bulk. Similar patterns have been found for the other waste bodies in this study as well.

In 2018 a number of samples were taken from Braambergen 11N while drilling holes for gas injection wells in the waste body. These solid samples were sent to the laboratory for further analysis. The chloride measurements for these samples ranged from 287 to 540 mg/kg dry waste. Assuming an average waste body height of about 10 m and a dry bulk density of 1,600 kg/m^3 this would imply values ranging from 4,592 to 8,640 g/m^2. The values in Figure 10 range from 3,560 to 5,436 g/m^2. Although these numbers should be considered with care, because of the uncertainties related to the total mass of waste and water content, the magnitude and range of the values are however, very similar.

These results indicate that our approach provides insight in the mass controlling long-term emissions from waste bodies. The total mass in Figure 10 shows the emission potential of this waste body as function of time. This mass controls the concentration in the leachate and as no new mass is added to the waste body it also will control future emissions.

5. Implications for Managing Landfill After Care

The model and parameter inference approach based on life-expectancy modeling is a viable approach to describe measured time series of leachate production rates and leachate concentration dynamics. Because the parameter inference approach provides us with uncertainty estimates, we also obtain insight in the uncertainty of the simulated values. The inferred parameters allow us to understand the uncertainty in the parameters controlling the emission of solutes from the waste body. The model allows us to make sense of the measured variations in leachate concentration data without having the need to smooth the data.

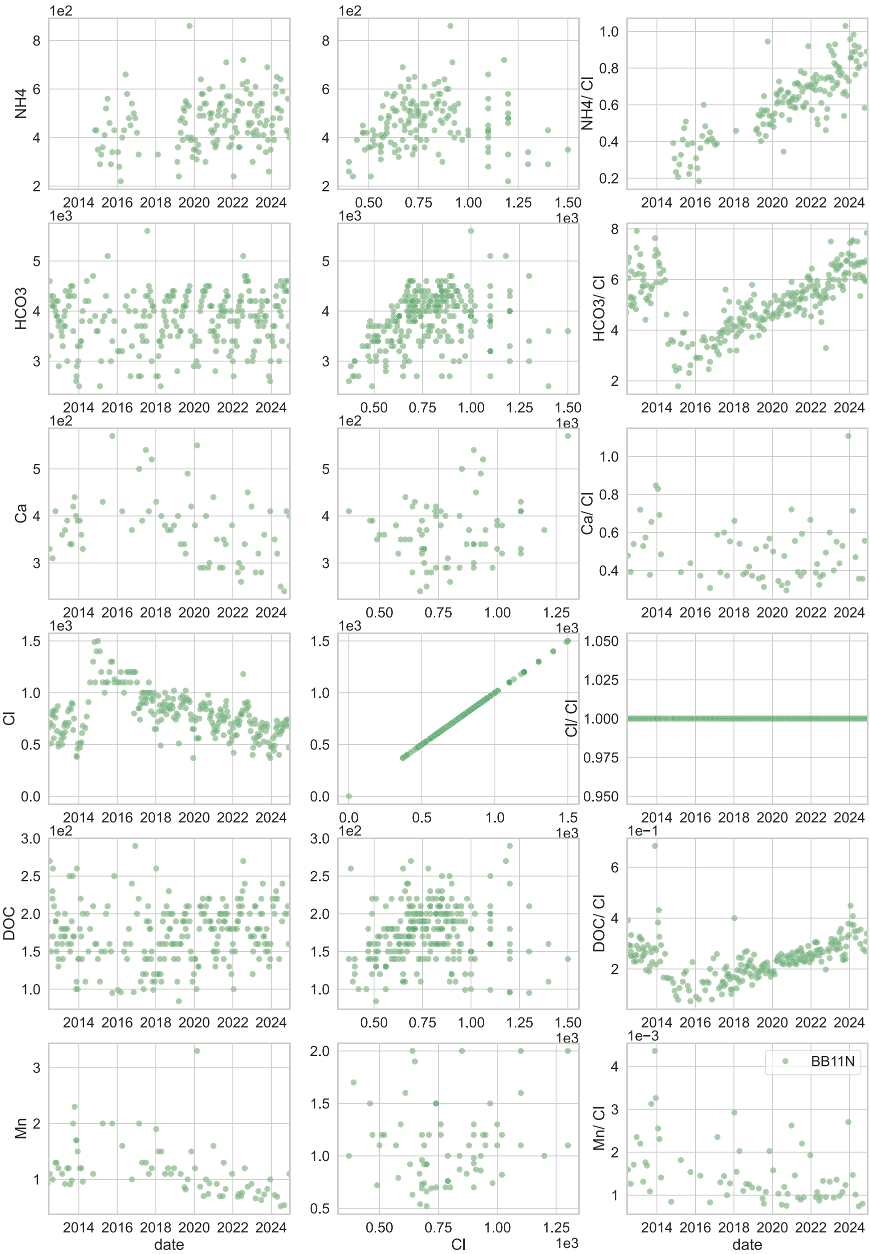

Figure 11. Time series of reactive parameter concentrations in the leachate of Braambergen 11N. The left columns shows the measured time series, the middle column shows the correlation with the measured chloride concentrations and the right column shows the time series of concentrations normalized to chloride.

An important result is that this approach can provide minimum and maximum estimates of leachate concentrations over time which can be used to assess the necessity of landfill after-care measures. In addition the inferred parameter distributions allow for an estimate of amount of mass in the waste body that is controlling the long-term leachate concentrations. This information is essential for understanding long term future risk associated with leachate emissions.

The approach we present currently only estimates the water balance and the emission of conservative solutes such as chloride which is a necessary first step to assess the emission of reactive compounds. We illustrate this in Figure 11 where time series of a selection of reactive parameters are shown. Normalizing all measurements by the chloride concentrations will illustrate the impact of dilution by preferential flow. If dilution has a significant impact, normalizing by chloride will remove a lot of variation from the time series. This is clearly seen for the time series of ammonium (NH4), bicarbonate (HCO3), and dissolved organic carbon (DOC) in the right column in Figure 11. A strong indication of non-conservative behavior is the increasing trend in these three time series. For calcium (Ca) and manganese (Mn) the behavior is different, the impact of dilution is much less clear.

The non-conservative behavior of these parameters is controlled by a multitude of processes. NH4, DOC and HCO3 are to a large extent controlled by the hydrolysis of solid organic matter in the waste body, where the dissolved compounds are further degraded under a range of redox conditions present in the waste body. Since 2018, the Braambergen 11N waste body has been aerated by the landfill operator by over extracting landfill gas from the waste body which effectively pulls in oxygen from the ambient air. As a result, in part of the waste body aerobic degradation can occur, leading to a significant increase in the CO_2 pressure. The behavior of HCO3 is also impacted by dissolution and precipitation of a wide range of carbonate minerals which are strongly influenced by the partial CO_2-pressure in the waste body. In anaerobic landfills the partial CO_2-pressure is in the order of 0.5 bar and in aerobic parts of the waste body it can be higher. Pumping of leachate from the leachate system leads to a large amount of turbulence in the pump-pit causing the leachate to mix with the ambient air, thus rapidly lowering the CO_2-pressure, which causes a significant precipitation of carbonate minerals in the pump-pit. This is probably the reason why the calcium concentrations do not seem to be influenced by dilution, they are most likely controlled by the solubility of calcium carbonate in the pump-pit. Manganese is a very redox sensitive parameter and it is soluble under anaerobic conditions. The time series of chloride normalized Mn concentrations implies that leaching is to a certain degree conservative.

Clearly, assessing emission potential from time series of reactive parameters is not straightforward. However, knowing the impact of dilution after assessing the water balance and the emission of chloride provide insights in the net effect of hydrolysis and degradation on the concentration of for example, NH4. Such insights will guide the development of reactive transport models with which we can assess the long term behavior of chemical compounds distributed over the solid mineral, solid organic, liquid and gas phases present in the waste body. The models will need to include processes such as dissolution and precipitation of minerals, hydrolysis of organic matter, sorption to reactive surfaces and the biological degradation of organic compounds under a wide range redox conditions. Such reactive transport models need to include both transport with the flowing water, as well as transport of the landfill gas migrating through the waste body.

The results presented in this paper clearly demonstrate that concentrations of chloride in leachate are dominated by dilution with water infiltrating from the cover layer and quickly moving via preferential path ways to the drainage system. Transport of mass from the bulk of the waste body to the drainage system is a relatively slow process. Dilution is a dominating mechanism reducing the actual leachate concentrations compared with the solute concentrations present in the waste body.

Environmental protection criteria are related to the actual concentrations in the leachate and if these are consistently below accepted values, landfill after care can be reduced (Brand et al., 2016; Dijkstra et al., 2018). Quantification of the emission potential is an essential step in assessing if the leachate concentrations comply with environmental protection criteria under a wide range of scenarios under the constraint of the boundary conditions close to the ones used for quantifying the model parameters.

Enhancing preferential flow by engineered measures in the waste body, may reduce these concentrations even further because the amount of water being stored in the bulk will decrease. This then leads to a decrease in the release rate from the bulk because of a decrease in storage. However the decrease in emission potential will slow down as well. When relying on such engineered measures it is important to ensure that the engineered solutions are robust and remain operational over extended periods of time under a range of varying boundary conditions.

Extreme scenarios such as flooding of the landfill may have to be taken in to account as well as these may lead to enhanced leaching (Laner et al., 2009; Nicholls et al., 2021). Emission potential estimates obtained by applying the methods proposed in this paper are a good starting point in evaluating the consequences of such extreme events.

This methodology requires long-term time series of leachate production rates and leachate concentration values in order to infer the parameters. Leachate production rates and leachate concentrations are parameters that landfill are obliged to measure in order to be compliant with the regulations. For the Wieringermeer and Braambergen landfills the landfill operator, Afvalzorg, did increase the measurement frequency significantly. When applying the approach to new data sets one can start using the posterior distributions obtained published with this paper, instead of starting with fresh uninformative priors.

6. Summary and Conclusions

A model has been developed for simulating leachate production rates and leachate concentrations using a mass balance approach combined with a stochastic travel time approach based on life time expectancies. The model is one dimensional and consists of two layers. The first layer is a reservoir model for the cover layer, the second layer a stochastic life expectancy model. The model is driven by measured rainfall and potential evapotranspiration and is calibrated using measured leachate production rates and leachate concentrations. Posterior parameter distributions are inferred using a Bayesian MCMC approach implemented in PyDREAM (Shockley et al., 2018) where the objective functions are based on the generalized likelihood model of Schoups and Vrugt (2010). The model has been applied to analyze data sets obtained from two different landfills and a total of four waste bodies.

We also propose to use emission potential as a term to describe the amount of mass that can be released from the waste body under realistic meteorological conditions. This emission potential is the source term for a conservative solute (chloride) in a modeling framework that can describe measured leachate flux and leachate concentration. Quantifying the water balance and conservative leaching is a necessary first step before evaluating the leaching of more reactive compounds.

The model with posterior parameter distributions can describe the measured time series of leachate production rates and leachate chloride concentrations. In addition uncertainty bandwidths using inferred measurement errors can be determined as well. Model simulations can be carried out to extrapolate leachate production and leachate concentration in to the future allowing for assessment of the future development of concentration and leachate production volume.

The hydrological parameters, travel time distributions and stagnant zone properties derived with the approach presented in this paper form the basis of a reactive transport model setup in which both conservative and reactive substances can be estimated simultaneously.

The results indicate that the waste bodies that have been studied, have reached seasonal steady state until 2022, where total water storage in the waste body oscillates around a constant value. From 2022 onward, the measured cumulative leachate production is larger than simulated. This implies that using time independent life-expectancy probability distributions is too strict.

The oscillations in leachate production rates and leachate concentrations are controlled by infiltration rates from the cover layer and the life time expectancy distribution. Leachate concentrations depend strongly on the simulated base flow which is controlled by the storage in the bulk of the waste body where life expectancy of water is longer than 1,825 days.

Emission potential is a combination of the total mass present in the waste body and the expected future behavior of the base flow. The model can be used to plot the future development of the two parameters controlling leachate concentrations being total solute mass and total water storage for different climate forcing scenarios. The total mass is a quantification of the emission potential. The results of such simulations can be used to assess different landfill after care scenarios and therefore it allows us to estimate leachate concentrations and the expected emission over a predefined time period.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

The software and data including documentation used for generating the results in this paper are available on the 4TU. ResearchData repository (Heimovaara & Wang, 2024): https://doi.org/10.4121/ade0d39f-8b07-4e48-8726-f900cf27141e.

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