LONG-TERM BEHAVIOR OF MUNICIPAL SOLID WASTE LANDFILLS

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(Received September 1988)

A method is presented to predict the long-term behavior of element concentrations (non-metals and metals) in the leachate of a municipal solid waste (MSW) landfill. It is based on water flux and concentration measurements in leachates over one year, analysis of drilled cores from MSW landfills and leaching experiments with these samples . A mathematical model is developed to predict the further evolution of annual flux-weighted mean element concentrations in leachates after the "intensive reactor phase", i.e. after the gas production has dropped to a very low level. The results show that the organic components are the most important substances to control until the leachate is compatible with the environment. This state of low emissions, the so-called "final storage quality", will take many centuries to be achieved in a moderate climate.

Key Words—Leachate, solid waste, elements, organic emission, final storage, drilled cores, modeling, Switzerland.

1. Introduction

In the present work, a municipal solid waste (MSW) landfill is regarded as a treatment facility whose objective is to obtain a residual solid of a "final storage quality", one which does not adversely affect the environment for long periods.

The important production of fuel gas in a controlled MSW landfill lasts about one to two decades (Ehrig 1986, Stegmann 1978/1979) . Pollutant transport by gas is negligible after this period except for laterally migrating methane which is both explosive and a possible cause of anaerobic zones in the soil for some time after useful production is completed . The leachate, however, remains the most important flux impacting the environment and must be collected for treatment. As a first approximation, a landfill, whose leachate is compatible with the environment, is defined in the present paper as a final storage.

Records of controlled landfills, i.e. with known input and output fluxes, have only been kept for about 20 years . The existing controlled landfills are still in the first phase (Fig. 1), here called the "intensive reactor phase", or at the end of it (Baccini et al. 1989). Their chemical behaviour can be most accurately assessed in this period (Baccini et al. 1987) . However the long-term behavior of MSW landfills is not known from experience . Even the oldest controlled landfill has not yet reached the final storage quality and the leachates have to be treated until the final storage quality is reached.

The crucial questions to be answered are as follows:

What is the further evolution of the leachate composition of a MSW landfill? When will MSW landfills reach the final storage quality?

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Fig. 1. Schematic illustration of the course of concentrations in gas and leachate from a MSW landfill as a function of time. Symbols are listed in nomenclature.

In the present paper, a rough estimation of the further evolution of some element concentrations (indicator parameters) in the leachate of a MSW landfill is made as a first attempt. For this purpose, a simple mathematical model is used, which is based on data from the intensive reactor phase of a MSW landfill and on data from leaching experiments of drilled core samples of the same landfill . The results are then used to assess the time required to reach the chosen quality standards, which are based on Swiss quality standards for running water (1975) .

2. Methods

2.1 Concept

The long-term emission potential of a landfill is determined by the further evolution of leachate water fluxes and metal and non-metal concentrations in leachates . For this purpose, the non-metals carbon, nitrogen, phosphorus and chlorine, and the metals iron, copper, zinc, lead and cadmium are chosen as indicator elements . The fluxes of these elements in leachates are then used to assess the further evolution of material fluxes from MSW landfills.

The annual leachate/precipitation ratio in the chosen landfills is approximately 0 .2 at the start and reaches a value of ca. 0.4 after filling is stopped (Baccini et al. 1987). No special surface treatment to divert precipitation was reported for these landfills . It can be assumed that the water content of the landfill is hydrodynamically stabilized after this time. This hydraulic regime is typical for a MSW landfill in moderate climates and a specific area charge of about 20×10^3 kg m⁻². Assuming a more or less constant annual precipitation, the annual leachate flux also remains constant after the intensive reactor phase .

The long-term behavior of a MSW landfill is assessed by extrapolating the results from observed fluxes in existing landfills . However the processes during and after the intensive reactor phase are not necessarily equal . The high gas production can accelerate the export of some elements . Nearly 25% of the organic carbon is exported by the output fluxes, predominantly in the landfill gas during the intensive reactor phase. More than 90% of the other non-metals and more than 99 .9% of the metals are still found in the residual solid at the end of the intensive reactor phase (Baccini et al. 1987). The lot

> Further remobilization processes depend on various factors, such as pH, redox conditions, salt concentrations, complexing agents, organic solvents etc. (Ham *et al.*) 1979, Stanforth et al. 1979, Förstner et al. 1988). These processes cannot be easily simulated by laboratory experiments or by lysimeters . As a first approximation it is postulated that:

the concentrations in leachates decrease exponentially with a first order rate ; the types of remobilization processes determining the concentration decrease in leachates during the predicted time period do not alter.

It is then possible to estimate the behaviour of element fluxes in a limited period (from $10¹$ to $10³$ years) on the basis of the following assumptions:

- (1) the pH-value of the leachate will not drop below 7 in the predicted time period;
- (2) other boundary conditions, such as salt concentrations, complexing agents etc . stay more or less constant i.e. they change only within an order of magnitude.

These assumptions will be commented on in section 3.

2.2 Mathematical model

A MSW landfill is a half-continuous fixed bed reactor . The fixed bed consists of deposited MSW. Water is the other input which enters the reactor continuously by precipitation . Chemical engineering methods can be applied to this reactor by introducing some simplifications (Levenspiel 1962) . For this purpose, the landfill is considered as a homogeneous reactor without concentration gradients in it . Thus, the mass balance for an element is given by equation (1) .*

 $Input + remobilization = output$ $I+M\times r=c\times V$

where I is the input of element by precipitation (mg year⁻¹), M is the amount of MSW (kg), which is assumed to be constant after the intensive reactor phase, r is the remobilization rate of the element (mg kg⁻¹ year⁻¹), c is the annual flux-weighted mean concentration of the element (kg 1^{-1}) and V is the annual mean leachate flux rate (1) γ ear⁻¹), which is assumed to be constant after the intensive reactor phase (see Section 2.1). The input I is practically 0 after the intensive reactor phase, because the contributions to the element fluxes from precipitation are negligible . Thus,

 $M \times r = c \times V$

(2)

^{*}A full list of symbols is given on page 56

For a first order remobilization rate in a homogenous reactor: 46
For a first o

$$
r = -\frac{\mathrm{d}m}{\mathrm{d}t} = k \times m \tag{3}
$$

where m is the concentration of the element in the MSW that can be mobilized (mg kg^{-1} , t is the time after the intensive reactor phase (year) and k is the remobilization rate constant (year⁻¹). Provided that k is time independent, equation (4) results by solution of the differential equation (3). 46
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For a first order remobilization rate in a homogenous reactor:
 $r = -\frac{dm}{dt} = k \times m$

(3)

where *m* is the concentration of the element in the MSW that can be mobilized ting

kg '*k*, *i* s

$$
m = m_0 \times e^{-kt} \tag{4}
$$

where $m₀$ is the initial concentration of the element in MSW that can be mobilized. The choice of time-independent k is based on the assumptions mentioned in section 2.1.

Hence, combining equations (2) , (3) and (4) leads to the following equation:

$$
c = \frac{kMm_{\theta}}{V} \times e^{-kt} \tag{5}
$$

The initial condition is as follows:

$$
t = 0; c = c_0 = \frac{k M m_0}{V}
$$
\n
$$
\tag{6}
$$

where c_{θ} is the annual flux-weighted mean concentration of the element in the leachate at the end of the intensive reactor phase (mg 1^{-1}). Thus.

$$
c = c_0 \times e^{-\frac{V c_0}{M m_0}t}
$$
\n⁽⁷⁾

Equation (7) gives the annual flux-weighted mean concentration c of the element in the leachate as a function of time. In order to determine the time t_F required to reach the final storage quality, equation (7) is transformed to equation (8).

$$
t_E = \frac{Mm_0}{Vc_0} \ln \frac{c_0}{c_E} \tag{8}
$$

where c_E is the concentration of the element which is compatible with the environment (see sections 1 and 4) .

The terms M, V and c_0 are measured, c_E is chosen on the basis of Swiss standards for running water, and $m₀$ is assessed by laboratory experiments. Residual solids, which are obtained from cores drilled in MSW landfills, are used for this purpose. The part which can be mobilized is determined by consecutive leaching experiments with the drilled core samples in the laboratory.

2.3 Experiments

2.3 .1 Sampling

Drilling was carried out in the MSW landfills in the area of Uttigen, Switzerland, where water and element balances were studied over one year (Baccini et al. 1987). Residual

solid samples of 50-100 kg were obtained from different layers. Five well mixed samples of 8-12 kg from every layer were immediately transported to the laboratory (Obrist 1986). After drying at 105° C the samples were sorted by hand to give the following components: metals; stones; glass; plastics; wood and paper and residue. The residue was ground in a beater mill and screened with a sieve. The resulting fraction $(< 0.5$ mm) was used for all leaching experiments.

2.3 .2 Leaching with water

Thirty grams of pulverized sample were mixed in an Erlenmeyer flask with 300 ml distilled water, the flask was covered with parafilm and rotary-type extracted at 150 r.p.m. for 0.5 h. The suspension was filtered (membrane filter $< 0.45 \mu$) and the extract was analysed. The filtration residue was leached again with distilled water under the same conditions for 5 h. This consecutive leaching procedure was carried out for a total of four times eventually giving four consecutive extracts of the same sample with leaching times of 0.5, 5, 50 and 288 h. The following parameters were analysed in the extracts: dissolved organic carbon (DOC), nitrogen, phosphorus, chlorine, iron, copper, zinc, lead and cadmium.

2.3 .3 Analytical techniques

The element concentrations in the aqueous extracts were determined as follows : DOC, determined with DOHRMANN DC 80; nitrogen, determined as the sum of NH_4^+ – N and DON (dissolved organic nitrogen) by the Kjeldahl method. Nitrate was negligible; phosphorus, determined as phosphate by the method of molybdenum blue after digestion in potassium peroxide sulfate solution; chlorine, determined as Cl^{-1} with ionsensitive electrode. Chlorine in chlorinated hydrocarbons was negligible and metals, determined by atomic absorption spectroscopy after digestion in aqua regia .

The element concentrations in the residue were determined as follows: total carbon, determined with CARLO ERBA 1500; total inorganic carbon, determined by the coulometric method; total nitrogen, determined with CARLO ERBA 1500; total sulfur, determined with CARLO ERBA 1500; phosphorus, determined by the method of molybdenum blue after digestion in aqua regia; metals, determined by atomic absorption spectroscopy after digestion in aqua regia .

2.3.4 Alkalinity

The alkalinity of the residue, defined operationally by the amount of acid to reach pH 7, was determined in 11 parallel batch titrations. The pH of the suspensions of 1 gram pulverized residue in 100 ml aqueous solutions with increasing aliquots of $HNO₃$ was measured after 48 h of rotary-type shaking (150 r.p.m.) in an Erlenmeyer flask.

3. Results and discussion

The mass distribution between the sorted fractions of two drilled core samples are presented in Table 1. The age of the samples were determined by the input records of the landfill company and by the dates on newspapers and magazines found in the drilled cores (Obrist 1986). The residue of sample 1 is 66% and the one of sample 2 is 51% in mass. As this fraction has the highest surface/volume ratio, it is the most important fraction in the mobilization of elements in the landfill . Therefore the leaching experiments are carried out with this fraction . It can be assumed that the contributions from

the other fractions are negligible compared with the residue . Because of this assumption, the amounts of elements mobilized in the leaching experiments are smaller than the amounts which could be mobilized theoretically in landfills . Consequently, the assessment of mobile parts of elements is probably underestimated by the methods applied in this paper.

Element concentrations in the residue are presented in Table 2 . Both samples have similar compositions, i.e. the concentrations of the non-metals do not differ significantly and differences in the metal concentrations are within an order of magnitude .

In Fig. 2 the concentrations of the non-metals carbon (dissolved organic carbon), nitrogen (Kjeldahl-nitrogen), phosphorus and chlorine are given as a function of time in the consecutive leaching experiments . There are significant differences between the two samples. But there are also differences between the two leaching experiments of the same sample (experiment 1 and 2). Sample 2 is mobilizing less, even though the total element

ı r ı ٠

Chemical composition of the residues of drilled core samples ; mean values and standard deviation of mean values (S.D.)

ND, not determined.

Fig. 2. Concentrations of some non-metals in aqueous extracts of the consecutive leaching experiments as a function of the leaching time; \Box , sample 1, leaching experiment 1; \Diamond , sample 1, leaching experiment 2; \Diamond , sample 2, leaching experiment 1; \blacksquare , sample 2, leaching experiment 2.

concentration in the solid does not differ from sample I (Table 2) . In general the concentrations in the first extract (after 0.5 h) are much higher than the concentrations in the subsequent extracts . It follows that the main fraction to be mobilized is found in the first extract.

Figure 3 gives the concentrations of the metals iron, copper, zinc, lead and cadmium in the extracts . The copper and zinc concentrations decrease with increasing extraction . The iron and lead concentrations in the second extract are nearly equal to or higher than the ones in the first extract . The increase of the cadmium concentration is not significant because of higher analytical errors in these concentration ranges.

The element concentrations, m_o , in MSW that can be mobilized are determined by addition of the mobilized amounts in the four consecutive extracts of each leaching experiment. Again this procedure can lead to an underestimation of the total mobilized fraction since the experiment does not guarantee that further extractions give negligible contributions . The sum of the water volumes applied in the four extractions corresponds to a leachate volume of 2000 years for a municipal solid waste landfill with 20 g annual leachate per kg MSW and year (Baccini et al. 1987).

In Table 3 the mobilized quantities in the residue are compared with total concentrations . The latter are compared with the mean concentrations found in MSW in Switzerland, which are determined by several mass balance studies of MSW incinerators (Brunner & Ernst 1986), from MSW compost data (W. Obrist, personal communication 1985) . All concentrations are given per unit mass MSW . There are two reasons for the significantly lower carbon contents in the residue with respect to the MSW . First, a fraction (about 25%) has already been exported as CO_2 and CH_4 during the intensive reactor phase (see section 2.1). Second the fractions "plastics" and "wood and paper", both important carriers of organic carbon, were separated (see Table 1) . Consequently the other non-metals also show lower concentrations in the residues than in fresh MSW .

Fig . 3 . Concentrations of some metals in aqueous extracts of the consecutive leaching experiments as a function of the leaching time. \Box , sample 1, leaching experiment 1; \bigcirc , sample 1, leaching experiment 2; \Diamond . sample 2, leaching experiment 1; \blacksquare , sample 2, leaching experiment 2.

Sorting out metals (Table I) can explain the reduction of the iron concentration . It did not significantly influence copper and zinc . The lower cadmium contents in the residues could be a result of the separation of cadmium containing plastics (PVC), ceramics and metals (pigments) . In conclusion it can be stated that the chosen residues are a reasonable first approximation of MSW at the end of the intensive reactor phase .

The composition of the residues and the leachability of elements show a certain heterogeneity. Consequently, the four leaching experiments do not give enough data to justify a mean value for the term m_0 in equation (7) from a statistical point of view. Therefore, the observed range (minimum to maximum value) is chosen and presented in the second column of Table 4 . For organic carbon a further transfer to inorganic carbon by microbiological activity after the intensive reactor phase is not considered, a process which could lower the $m₀$ -values for DOC. It would, however, be compensated for by a further decomposition of the non-mobile organic compounds in the residue and the components "wood and paper" and maybe even "plastics" to mobile species . Therefore, the m_0 range for C_{org} is not overestimated.

TABLE ³

Concentration of elements in MSW, relative amounts of elements in the residue and sum of the quantities mobilized by four consecutive extractions per mass unit MSW

* Brunner & Ernst 1987, Obrist pers . comm. 1985 .

t Determined by the difference of total carbon and total inorganic carbon .

ND, not determined.

TABLE 4

Element concentration, $m₀$, in MSW that can be mobilized; annual flux-weighted mean element concentration, c_0 , in the leachate at the end of the intensive reactor phase; chosen quality standard, c_E , for the leachate (Swiss ordinance 1975); and time, t_{FS} , required to reach the final storage quality with respect to the leachate

* Percent of total concentration in MSW.

 \dagger Sum of NH₃ + NH₄⁺.

 \ddagger Tenfold value of upper phosphate level in oligotropic lakes.

§ Quality criteria for sulfate.

 $V/M = 0.02$ 1 kg⁻¹ MSW years⁻¹.

ND, not determined.

The further evolution of element concentrations c in leachates can be calculated with equation (7). The annual flux-weighted mean element concentrations, c_0 , in the leachate at the end of the intensive reactor phase are given in column 3 of Table 4 . They were taken from field experiments presented in an earlier paper (Baccini *et al.* 1987). V and M were measured in the same landfill. V/M is assumed to be constant because the landfill is hydrodynamically stabilized after the filling is stopped and the gas production is very low after the intensive reactor phase so that the decrease in M is negligible (Baccini et al. 1987). The errors introduced with $c₀$ and V/M are much smaller than the error in the assessment of m_o .

The time period for the assessment is limited to 2000 years . For such a long period the following question has to be answered: Is the most important assumption probable, that the master variable pH stays above 7 to ensure a more or less constant physical and chemical condition in the distribution of species between the solid and the solvent phase, from a thermodynamic point of view?

The titration curves of the alkalinity measurements (see also section 2.3.4) are given in Figure 4. The pH of the leachates formed in the extraction experiments is about 8 .3 . The alkalinity is approximately 6 mole kg^{-1} residue for both samples. The organic carbon concentrations in the drilled cores after the intensive reactor phase are less than 100 g kg⁻ (Obrist 1986). Provided that 50% of this carbon (worst case) can be oxidized to CO, in the next 2000 years, less than 4 moles of weak acids will then be produced. The proton flux by precipitation (50 g precipitation with a pH of 4 kg^{-1} MSW year⁻¹) is less than 10^{-5} mole kg⁻¹ residue year⁻¹. This proton source would be still two orders of magnitude lower than the proton potential of the residue itself in a time period of 2000 years . It follows that the assumption of maintaining alkaline conditions for 2000 years is plausible . Consequently, a higher remobilization rate of some metals due to lower pH is not expected, at least for many centuries.

Another assumption is that salt concentrations are constant during the estimated period. Ionic strength is between 10^{-1} mole 1^{-1} and 10^{-2} mole 1^{-1} in the leachate during the intensive reactor phase. It is higher than 5×10^{-3} mole 1^{-1} after the intensive reactor phase due to dissolved carbon dioxide (Stumm $\&$ Morgan 1970). The change is within one order of magnitude .

Fig. 4. Titration curves of the residue, ■, sample 1; \Box , sample 2.

The element concentrations decrease in the consecutive extracts of a leaching experiment. This points to a remobilization process of higher than zero-order. Therefore, a decrease in concentrations in the leachate with a first-order kinetic is plausible as a first approximation.

> Other boundary conditions could be more or less stimulated by leaching experiments . As the results are discussed in orders of magnitude, this simulation is a reasonable approach.

> The annual mean concentration c as a function of time is calculated for each element, according to equation (7) and the data for $m₀$ and $c₀$ given in Table 4. The results are presented graphically in Figs 5 and 6 . The two curves are due to the two extreme values of m_o . The range between them gives the most probable course of concentration in the leachate. The DOC concentration in the leachate reaches a value of about $100 \text{ mg } 1^{-1}$ after many hundreds of years, the nitrogen and chlorine concentrations decrease to low levels within decades . Centuries are needed to halve the concentrations of the other elements.

> In order to answer the crucial question about the time required to reach the final storage quality, quality standards c_E , i.e. concentrations compatible with the environment, have to be chosen. They are given in column 4 of Table 4. For organic carbon (DOC), nitrogen (ammonium) and the metals the tenfold Swiss quality standards of running water (Swiss Ordinance 1975) are taken . For fluorine (fluoride), sulfur (sulfate) and chlorine (chloride) the original values of the quality standards were chosen . These ions generally show a higher mobility in natural or synthetic liners . For phosphorus the tenfold value of the upper phosphate level in oligotrophic lakes (0.04 mg l^{-1}) is taken.

> The time t_E required to reach the final storage quality can be calculated with equation (8). The time t_{FS} is the total time from the start to the final storage quality and is equal to t_F + 10 years, where 10 years is chosen for the intensive reactor phase. The results for t_{FS}

Fig . 5 . Evolution of some non-metal concentrations in the leachate of a MSW landfill after the intensive reactor phase .

Fig . 6 . Evolution of some metal concentrations in the leachate of a MSW landfill after the intensive reactor phase.

are presented in column 5 of Table 4. Again the ranges given are due to the corresponding ranges for m_p . Fluorine, sulfur and all metals can reach final storage quality within the intensive reactor phase, due to the fact that c_0 and c_E are of the same order of magnitude (Table 4) . Nitrogen needs at least several decades . The longest period is needed for chlorine, phosphorus and organic carbon, i.e. at least one to several centuries. This order of magnitude is not questioned by the rough estimation of m_q .

All assumptions taken were very conservative with respect to the potential of the mobile fraction. Therefore the calculated values for c in Figures 5 and 6 are somewhat underestimated. Consequently the values for t_{FS} could only be lowered if one accepts higher concentrations c_E for the quality standards. Our knowledge is still very incomplete for the bulk of organic substances leaving a MSW landfill . Thousands of xenobiotics are found in various products of MSW . Some of them may be decomposed within the landfill, others may be stable but non-hazardous to the receiving aquatic system. However, a third group may be transformed into toxic compounds. Since we know so little about the chemical composition, the chemodynamics and long-term behavior of these classes of substances, the quality standard should be set at a low-level concentration for the bulk parameter DOC .

Fig. 7. Comparison of times required to reach the final storage quality. ■, Calculated by the method presented in this paper; \mathcal{W} , calculated by the method presented in Belevi & Baccini 1987.

The estimation of m_0 is the most speculative part of the assessment procedure. Another method for the estimation of $m₀$ is based on the chemical speciation of the elements in MSW. This method is described elsewhere (Belevi & Baccini 1987) . A comparison of the time ranges t_{FS} for organic carbon, nitrogen, phosphorous and chlorine, which are calculated by both methods, are illustrated in Fig . 7 . Even though the estimations of m_0 are independent of each other, the resulting times t_{FS} are of the same order of magnitude.

4. Conclusions

The assessment of long-term element fluxes from MSW landfill leachates leads to the following conclusions:

After the intensive reactor phase, i.e. after 10 to 20 years (Fig. 1), the organic compounds are the most important substances to control . If a quality standard near surface water properties (within an order of magnitude) has to be reached, treatment of the leachates for at least several centuries will be necessary.

If the objective of a waste management policy is to ensure that future generations are forced to deal with the wastes of their ancestors, each generation must transform its wastes to final storage quality. This objective cannot be reached for the MSW landfills, their leachates, for example, must be treated for much longer than a human generation, in order to avoid a harmful environmental impact.

The method for the assessment of the long-term course of leachate concentrations is applicable to other type of landfills (e .g . bottom ashes) . It is however a costly method, due to the fact that field experiments are needed to obtain accurate values for the initial concentrations c_q . Furthermore the accuracy of the mobile fraction m_q , the most difficult term to assess experimentally, depends on the quality of the samples (homogeneous and representative) and the quantity of leaching experiments .

Acknowledgments

The authors thank Walter Ryser of the AVAG company for providing the opportunity to study the landfills in Uttigen, Switzerland .

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Nomenclature

- \boldsymbol{c}
- c_{θ} annual flux-weighted mean element concentration in the leachate at the end of the intensive reactor phase (mg 1^{-1})
chosen quality standard for the leachate (Swiss ordinance 1975) (mm 1^{-1})
- $c_{\overline{k}}$
- I input of element by precipitation (mg year^{-1})
- k remobilization rate constant (year⁻¹)
- m element concentration in MSW that can be mobilized (mg kg⁻¹ MSW)
- $\frac{m_0}{M}$ initial element concentration in MSW that can be mobilized (mg kg^{-1} MSW)
- amount of MSW
- r annual mean remobilization rate (mg kg⁻¹ year⁻¹) time after the intensive reactor phase (year)
- t
- t_{FS} time required to reach the final storage quality with respect to the leachate (year)
- t_k time required to reach the final storage quality with respect to the leachate after the intensive reactor phase (year)
time from the start to the end of the intensive reactor phase (year)
- $\frac{t_R}{V}$
- annual mean leachate flux (1 year^{-1})