

An approach for estimation of contaminant release during utilization and disposal of municipal waste combustion residues

D.S. Kosson^{a,*}, H.A. van der Sloot^b, T.T. Eighmy^c

^a*Department of Chemical and Biochemical Engineering, Rutgers, The State University of New Jersey, Piscataway, NJ, USA*

^b*Energy Research Foundation, Petten, The Netherlands*

^c*Environmental Research Group, University of New Hampshire, Durham, NH, USA*

Received 20 December 1994; accepted 26 July 1996

Abstract

The use of appropriate leaching tests and leaching models can help to predict how constituents in municipal waste combustion residues will leach during either utilization or disposal scenarios. This paper presents a general approach for estimating constituent release from MWC residues under a variety of management scenarios through use of fundamental leaching, site specific design and regional climatic parameters. Leaching behavior is categorized as being controlled by either (i) constituent availability or solubility for percolation-dominated scenarios with loose granular residues, or, (ii) controlled by diffusion for flow around scenarios with compacted granular residues or monolithic products containing residues. Three broad scenarios involving either disposal or utilization are used to illustrate the approach. The scenarios are applied to bottom ash, combined ash and APC residue. In two specific cases pertinent to bottom ash utilization, field data are used to verify the approach. Field data are also used to verify the approach for disposal of combined ash. These methodologies hold promise for serving as a basis for evaluating and comparing potential environmental impacts from different management scenarios for combustion residues and for other waste materials.

Keywords: Ash; Leaching; Utilization; Incineration; Disposal; Use; Management

1. Introduction

Municipal waste combustion (MWC) is a component of integrated waste management in many countries. The primary reasons for MWC are to (i) reduce the mass and volume of waste requiring disposal; (ii) recover energy from waste; and, (iii) produce

* Corresponding author.

a more inert material for disposal or alternative management practices. MWC typically results in approximately 90% and 70% reduction in volume and mass, respectively, of municipal solid waste. MWC residues, frequently collectively referred to as 'ash' require either disposal or alternative management. This has resulted in development of several management strategies, including utilization of MWC residues as an aggregate in construction applications and disposal in monofills. Comparison of the various management strategies should include evaluation of the long-term impacts from each alternative. This requires methodology for estimating release of various residue constituents during different management scenarios over extended time frames. Current practice in most regulatory jurisdictions relies on evaluation based primarily on regulatory leaching tests or short-term field studies. The basic premise of this paper is to offer an alternative approach for estimating long-term constituent release from MWC residues. This alternative approach is illustrated for three different management scenarios for bottom ash, combined ash and air pollution control (APC) residues. Estimated releases then are compared to field observations for two bottom ash utilization scenarios under conditions similar to those assumed for development of the estimates. Release observations also are compared with field data for landfill disposal of combined ash.

2. MWC residues

Many different MWC residue streams with different characteristics can be produced based on the differences in combustion and APC system design, waste feed composition and operating conditions. This paper focuses on residues from European style mass burn facilities with semi-dry scrubbers and fabric filter particulate collection (mass burn/DS/FF) to illustrate methodology for estimating constituent release. A similar approach can be applied to residues from other facility configurations.

The principal residues generated from European style mass burn facilities with semi-dry scrubbers and fabric filter APC systems are: (i) grate ash; (ii) grate siftings; (iii) fly ash and (iv) APC residues. Grate ash is the material retained on the grate after combustion has been completed. This material most frequently is discharged from the grate into a quench tank and then removed via conveyor. Grate siftings is the material which falls through the grate during combustion. The quantity and characteristics of grate siftings can vary as a function of facility design, grate conditions during operation and waste feed composition. Grate siftings often are discharged into the quench tank with the grate ash. Grate ash and grate siftings are most often mixed together and collectively are referred to as 'bottom ash'. Fly ash is defined as the particulate entrained in the combustion gases which is either carried over from the primary combustion chamber or formed during cooling of combustion gases. Fly ash may be collected prior to acid gas scrubbing (usually in an electrostatic precipitator (ESP)) or included in the residues collected after scrubbing. APC residues include fly ash, dry or semi-dry scrubber residue, and particulate from the fabric filters; the use of the term 'APC residue' in this paper refers to mixture of these residues. The relative proportions of grate ash, grate siftings, APC residues is approximately 300, 5 and

37 kg/tonne municipal solid waste (MSW), respectively [1]. In the United States, bottom ash and APC residues are frequently mixed together and referred to as 'combined ash' which is most frequently disposed of in lined, dedicated landfills or landfill cells with leachate collection. In Europe and Canada, these waste streams are usually separated [1].

Bottom ash has physical and chemical properties that make it amenable for use as an aggregate substitute in construction applications. In Europe, bottom ash is used as compacted road base material; structural fill in wind barriers, sound barriers, and highway ramps; and in asphalt applications [1].

Utilization rates are approximately 90% for the Netherlands, 72% for Denmark, 50–60% for western Germany, 26% for Switzerland, and 64% for France. In the United States, bottom ash is being considered as an aggregate for use in bituminous pavement and Portland cement applications [2]. Use of combined ash as an aggregate substitute in asphalt, Portland cement concrete, and as final cover or gas venting layers at municipal solid waste landfills also is being considered in the United States. In the Netherlands, fly ash collected in ESPs is used as a fine aggregate filler in asphalt. In Europe and Canada, scrubber residue is generally treated for secure disposal and not considered for utilization [1].

Table 1 provides typical elemental composition of bottom ash, fly ash, APC residues and combined ash from a mass burn/DS/FF facility [1]. Bottom ash can be

Table 1
Typical ranges and values of total content for bottom ash, fly ash, APC residue and combined ash from mass burn MWC facilities. All units in (mg/kg)

Element	Bottom ash [1]	Fly ash [1]	APC residue [25]	Combined ash [25]
Al	52 000 [25]	na ^a	26 000	55 000
As	5–40	1–2	90	17
Ba	500–1800	30–80	320	750
Ca	50 000–90 000	50 000–100 000	290 000	120 000
Cd	2–25	100–300	140	31
Cl	1000–3000	30 000–50 000	170 000	28 000
Cr	200–1000	10–100	190	400
Cu	1200–2500	300–1000	500	1700
Fe	76 000 [25]	na	6200	67 000
Hg	0.5–1	1–20	9	3
K	7000–20 000	20 000–40 000	16 000	13 000
Mg	10 000–30 000	10 000–30 000	na	na
Mn	1100 [25]	na	420	2100
Mo	5–30	20–60	na	na
Na	20 000 [25]	na	20 000	21 000
Pb	1500–3000	4000–8000	3100	1000
SO ₄	12 000–30 000	30 000–90 000	6000	1400
Sb	30–200	150–500	1100	250
Zn	2000–4000	10 000–20 000	17 000	6100

^a na: Data not available.

classified as well graded sand and gravel, with a fines (<60 μm) content of up to 10%. Up to 20% can be oversized material (>10 cm) which frequently is separated from the bottom ash at the facility by coarse screening. APC residue is classified as a fine powder with 50% of its particle size being less than 80 μm .

The major elements (>10 000 mg/kg, listed in decreasing order of abundance) in bottom ash are O, Si, Fe, Ca, Al, Na, K and C (Table 1). These elements account for 80 to 90% of the weight of bottom ash. Their concentrations are similar to those seen in geologic materials. The minor elements (1000 to 10 000 mg/kg) include Mg, Ti, Cl, Mn, Ba, Zn, Cu, Pb, and Cr. The trace elements (<1000 mg/kg) in bottom ash include Sn, Sb, V, Mo, As, Se, Sr, Ni, Co, Ce, Ag, Hg, B, Br, F, and I. Although grate siftings comprise a small weight percentage of bottom ash (1–3%), they contribute a significant fraction of the elemental Pb, Al, Cu, and Zn to the bottom ash stream from mass burn facilities. The dissolvable solids content of bottom ash is between 3% and 14% and consists mostly of calcium and sodium salts, e.g., NaCl, CaCl₂, CaSO₄ [1].

The major elemental constituents (>10 000 mg/kg, listed in decreasing order of abundance) of fly ash are Si, Ca, Al, Cl, Na, K, S and Fe. Most elements are present as oxides, and O is the most abundant element in the fly ash. However, the presence of elemental Al has been observed in fly ash and may cause hydrogen evolution when the ash is contacted with water under natural, usually alkaline, conditions. Substantial amounts of Zn, Mg and Pb are also present. All these elements are present in concentrations exceeding 10 000 mg/kg. The minor elements Ti, Ba, Sn, Mn, and Cu are present in concentrations between 1000 and 10 000 mg/kg. The trace elements Cr, Sb, Cd, Sr, Ni, As, Ag, Co, V, Mo and Se, are present in concentrations less than 1000 mg/kg. The content of Hg in fly ash is normally less than 30 mg/kg. In addition, the water soluble fraction of fly ash is approximately 21–60 wt% [1].

The major elemental constituents (>10 000 mg/kg, listed in decreasing order of abundance) of semidry APC system residues are Ca, Cl, Si, Al, Na, K, S, Zn, Mg, Pb, Fe and O. CaCl₂ and CaSO₄ are present in elevated concentrations as a result of acid gas scrubbing. The minor elements Ti, Ba, Sn, Mn, and Cu are present in concentrations between 1000 mg/kg and 10 000 mg/kg. The trace elements Cr, Sb, Cd, Sr, Ni, As, Ag, Co, V, Mo and Se are present in concentrations less than 1000 mg/kg. The content of Hg in APC residue from semidry scrubber systems is normally less than 50 mg/kg. However, wet scrubber sludge may contain 500 to 1000 mg Hg/kg. APC residues are highly water-soluble with the water soluble fraction ranging between 27 and 65 wt% [1].

The composition of combined ash reflects the mixture of bottom ash and APC residue at a mass ratio of approximately 70:30, respectively. Fly ash included with the APC residue typically is 20% of the total quantity of residues produced. Frequently, a high degree of variability associated with sampling and analysis of bottom ash and combined ash will result in statistically indistinguishable analyses for several elements. This is a result of the intrinsic heterogeneity of these materials at a micro-scale. The primary differences between bottom ash and combined ash is that combined ash has elevated concentrations of Ca, Cl, S, Cd, Zn, and Hg, as well as TDS, relative to bottom ash.

Previous studies have identified Pb, Cd and total dissolved solids (TDS) as important constituents that may be leached from MWC residues, potentially causing detrimental environmental effects [2]. Copper and molybdenum also may be of concern in some aquatic environments [3]. Chloride is major component of the TDS originating from MWC residues and has been observed to contribute up to 50% of the TDS in leachates from MWC residue disposal [4]. Thus, Pb and Cl were selected to illustrate release estimates for this paper.

3. Leaching fundamentals

Currently, potential constituent release is most frequently evaluated based on single extraction leaching tests such as the Toxicity Characteristic Leaching Procedure (TCLP, United States) [5], DIN 38414 S4 (Germany) and AFNOR X-31-210 (France) [6, 7]. All of these are single batch extractions which use either acetic acid or distilled water as the extractant. In general, leaching tests can be classified into the following categories: (i) tests designed to simulate constituent release under specific environmental conditions (e.g., synthetic acid rain leach test or TCLP), (ii) sequential chemical extraction tests, or (iii) tests which assess fundamental leaching parameters. Tests that are designed to simulate release under specific environmental conditions are limited because they most often do not provide information on release over different time intervals or under environmental conditions different than the one being simulated. Sequential chemical extraction tests evaluate release based on extraction of the waste with a series of increasingly more aggressive extractants. This approach, originally compiled by Tessier et al. [8] was further adapted to wastes by Frazer and Lum [9]. Recent results have shown that resorption and reprecipitation reactions can dramatically alter the releases observed and thus invalidate the use of this approach [10, 11].

An alternative approach to estimating constituent release is through measurement of fundamental leaching parameters. The following fundamental parameters can be used as a foundation for estimating constituent release under a variety of environmental exposure conditions:

- Availability;
- Solubility; and,
- Apparent diffusion coefficient, tortuosity and chemical retention.

Each of these parameters are described in the discussion which follows.

3.1. Availability

Availability is defined as the maximum quantity or soluble fraction of a residue constituent that can be released into solution under aggressive leaching conditions [12]. These conditions, in theory, should provide an estimate of the maximum mass of material that could leach under a 1000 to 10000 yr time frame, except for mobile species such as highly soluble salts (e.g., sodium chloride) for which the availability can be reached in a matter of years. Under availability controlled conditions, the resulting solution (leachate) is at a concentration less than the saturation condition for the

element or species of interest. For example, availability in bottom ash typically excludes elemental species which are tightly bound in glassy matrices and in geologically stable mineral forms such as Si in SiO_2 (quartz), Ca in $\text{Ca}_2\text{Al}_2\text{SiO}_7$ (gehlenite), and Mg in $\text{MgCa}_2\text{Si}_2\text{O}_7$ (ackermanite). Thus, the availability of a specific element can be significantly less than the total content of that element (e.g., Pb) or may be approximately equal to the total content (e.g., Cl). Determination of availability, however, does not indicate whether or not this maximum quantity of a particular constituent will be released, or over what precise time interval the release will occur for the environmental exposure scenario of interest.

The following experimental conditions have been observed to result in availability controlled release for most elements and species of interest in MWC residues [1]:

- Liquid-to-solid (LS) ratios greater than 10 concurrent with pH less than 4;
- LS greater than 10 concurrent with near neutral pH and 0.2 molar citric acid; and,
- LS greater than 10 concurrent with near neutral pH and 0.016 molar EDTA;

All three of the above availability controlled leaching conditions result in the same quantitative release of a specific element or species for a specific ash sample [1]. The exception is Pb from APC residues, which often shows a maximum availability at pH greater than 11. However, none of these extraction conditions are intended to simulate actual field exposure scenarios. Two standardized leaching procedures which result in measurement of constituent availability are the Dutch Availability Leach Test (NEN 7341) [1] and the California Waste Extraction Test [13]. Availability leaching test results should be presented as the measured release (mg element leached/kg of material extracted).

Table 2 presents the availability of several elements and species from bottom ash, fly ash, APC residue and combined ash. The availability of specific elements or species of interest may be a small fraction of the total concentration present in the ash and may be significantly different for different ash types. For example, Pb availability in bottom ash, fly ash and APC residues is approximately 8%, 3% and 32%, respectively, of the total Pb content in the MWC residues. Conversely, Cl has high availabilities in all of the residue types. APC residues are significantly higher in Cd, Pb, Zn, Cl and SO_4 availability per unit mass of residue compared to bottom ash.

The relative amount of a specific element available for leaching as a function of ash type can be estimated based on the availability multiplied by the mass fraction of total quantity of residues that the particular ash type represents. For example:

- The total amount of Cl available for all residues is approximately 6000 g/tonne MSW of which 1 g/tonne MSW (0.02%) is present in grate siftings, 70 g/tonne MSW (1%) is present in grate ash, 3 g/tonne MSW (0.05%) is present in boiler ash and the balance, approximately 5900 g/tonne MSW (>98%) is present in APC residue.
- The total amount of Pb available for all residues is approximately 100 g/tonne MSW of which 50 g/tonne MSW (50%) is present in the grate siftings, 40 g/tonne MSW (40%) is present in the grate ash and, and 10 g/tonne MSW (10%) is present in the APC residues.

These results provide guidance on the relative potential environmental burden from each residue stream.

Table 2

Typical ranges and values of availability for bottom ash, fly ash, APC residue and combined ash from mass burn MWC facilities. All units in (mg/kg)

Element	Bottom ash [1]	Fly ash [1]	APC residue [25]	Combined ash [25]
Al	5600 [25]	na ^a	2900	4000
As	0.3–5	1–2	14	6
Ba	50–200	30–80	90	130
Ca	20 000–70 000	50 000–100 000	200 000	79 000
Cd	0.5–5	100–300	130	27
Cl	1000–6000	30 000–50 000	146 000	32 000
Cr	2–10	10–100	6	3
Cu	50–200	5–20	200	380
Fe	2700 [25]	na	30	670
Hg	0.01–0.1	1–3	17	0.6
K	1000–3000	10 000–25 000	14 000	5800
Mg	1000–3000	4000–15 000	4300	4700
Mn	220 [25]	na	64	680
Mo	1–4	1–4	11	2
Na	5600 [25]	na	14 000	5800
Pb	50–300	100–300	1000	na
SO ₄	8000–18 000	30 000–80 000	88 000	29 000
Sb	30 [25]	na	110	< 8
Zn	50–500	5000–8000	7900	2900

^a na: Data not available.

3.2. Solubility

The rate of constituent release is most frequently either solubility controlled or mass transfer controlled. Solubility control occurs when the solution in contact with a waste is saturated with respect to the constituent species of interest. This condition is prevalent at low LS (which typically occurs in the field) when percolation dominates water contact with contaminated solids. The solubility of most heavy metal cations (e.g., Pb, Cd, Zn, etc.) is strongly a function of solution equilibrium pH. Solubility of a particular element also can be increased by the presence of significant concentrations of complexing agents such as chloride, or acetate or reduced by the presence of a co-precipitating species such as sulfate or sulfide. Similarly, solubility can be increased during laboratory extraction procedures by the presence of these complexing agents or more aggressive chelating agents such as EDTA. Solubility typically is measured using a low LS (e.g., less than or equal to 5 l/kg) under equilibrium conditions. Two test methods which have been used to measure solubility as a function of solution pH are the pH-stat test [14] and the acid neutralization capacity leach test [15]. Solubility leaching results should be presented as solution concentration (mg/l) as a function of pH.

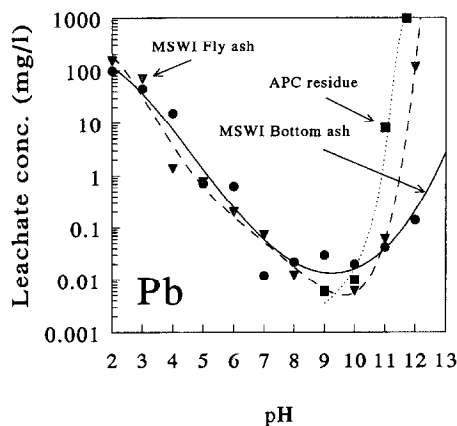


Fig. 1. The log-mean solubility of lead as a function of pH for bottom ash, APC residue and combined ash [1].

Fig. 1 presents the log-mean solubility of lead as a function of pH for bottom ash, APC residue and combined ash based on laboratory testing of a large number (>1000) samples of MWC residues [1]. Lead solubility is similar for all three residue types although the solubility of Pb from APC residue is slightly increased under alkaline conditions. These results indicate that lead solubility varies by three orders of magnitude as a function of solution pH. Minimum solubility is observed between pH 8 and 10.

Chloride is highly soluble with solubilities in neat aqueous solution of approximately 165 000, 216 000 and 475 000 mg Cl/l for KCl, NaCl and CaCl₂, respectively [16]. Solubilities can be increased relative to these values in solutions with other ions present and of high ionic strength. Chloride solubility also is relatively independent of pH and is released rapidly as a function of LS. Fig. 2 presents the leachate chloride concentration as a function of LS during laboratory column leaching tests carried out in accordance with NEN 7343 [17, 18]. Approximately 25% of the available chloride was released from bottom ash and fly ash at LS of 0.1 l/kg ash. Approximately 60% and 90% of the chloride availability was released by LS of 1 for bottom ash and fly ash, respectively. If 25% of the availability is used to estimate the initial concentration of chloride released during percolation controlled leaching, expected initial concentrations are approximately 7000, 150 000, 290 000 and 64 000 mg/l for bottom ash, fly ash, APC residue and combined ash, respectively. The initial concentrations of chloride observed during the laboratory column leaching tests were 5000 and 210 000 mg/l for bottom ash and fly ash, respectively. Field observations of bottom ash and combined ash leaching have indicated initial chloride concentrations ranging from 1400 to 1800 and 14 000 to 50 000 mg/l, respectively [19, 20]. These results primarily reflect flow heterogeneity (channeling and bypass) and that leachate produced in the field at LS less than 1 l/kg results from the displacement of pore water present during disposal and not infiltration which has percolated through the entire fill material.

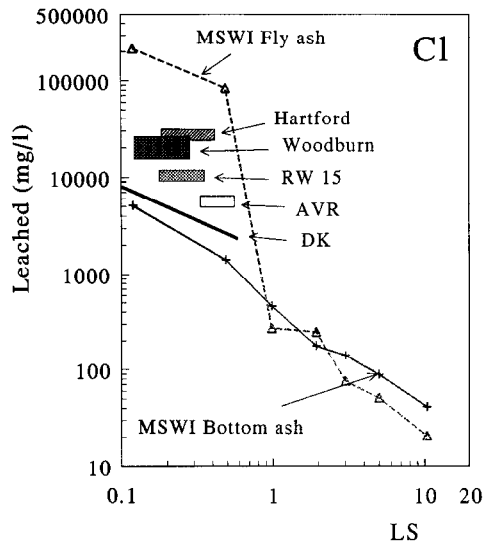


Fig. 2. Chloride leachate concentration as a function of LS during laboratory leaching tests for bottom ash and fly ash (MSWI Fly ash [18]; Woodburn, combined ash [19]; RW15, bottom ash [38]; AVR, bottom ash [18]; DK, bottom ash and fly ash [4]).

3.3. Diffusion

Mass transfer-controlled release occurs when the rate of constituent release from the solid matrix is the release limiting step and thus saturation or equilibrium with the leachate is not achieved. Constituent diffusion through the solid phase is usually the mass transfer controlling process. This condition is prevalent when MWC residues are monolithic in form (such as stabilized residues or residues incorporated into asphalt pavement) or, when granular materials are compacted to low permeability or overlain by an impermeable barrier.

The cumulative release of a diffusing constituent from a solid matrix, e.g. a monolithic solid or a compacted granular material, can be described by [21]:

$$M_t = 2\rho C_0 \left(\frac{Dt}{\pi} \right)^{0.5} \quad (1)$$

Eq. (1) assumes that the concentration of the diffusing constituent remains dilute at the surface boundary and that the concentration of the diffusing constituent at some distance from the surface boundary within the material does not change, e.g., depletion does not occur. Depletion occurs when the cumulative release of a very soluble component approaches the maximum leachable quantity, as determined by the availability leach test. The initial concentration of the diffusing element or species is equal to the availability of that component because the availability is the concentration which serves as the chemical 'driving force' for mass transfer. Use of the total elemental concentration as C_0 would result in overestimation of the cumulative

release or underestimation of the diffusion coefficient if that variable was being estimated from release data. Van der Sloot [22] reported that the observed diffusion coefficient can be divided into the free diffusion of the constituent of interest, a physical retention factor, and a chemical retention factor according to the relationship

$$D = \frac{D_{0,x}}{R\tau} \quad (2)$$

Physical retardation (τ) is a property of the solid matrix (either monolithic or compacted granular materials) and is independent of the diffusing element. It is equivalent to the tortuosity for aqueous phase diffusion for cases where the test sample remains physically intact (e.g., no significant cracking or disintegration). In these cases, tortuosity is an approximation of the ratio of the actual mean path length a species travels from within the monolith to the monolith surface for release to the mean direct geometric path length. Physical retardation values can be very high in hydrophobic matrices such as asphalt. For cases where the monolith does not maintain physical integrity, the physical retardation factor is a relative reference index for the degree of species retention within the matrix by physical encapsulation at the microscale.

The chemical retardation factor quantifies the decrease in the rate of the release of a component resulting from chemical interactions of the diffusing ion with the solid matrix by comparison to release of an inert species. The interactions typically involve sorption processes to surfaces and temporary surface-associated precipitation phenomena. Chemical retention is usually inversely related to the solubility of an element as a function of porewater pH within the solid matrix [14].

Diffusion coefficients, physical retardation factors and chemical retardation factors are calculated from data obtained during the monolith or compacted granular leach tests in conjunction with either the availability or total solid phase concentrations. ANS 16.1 [23] and NEN 7345 [24] are two experimental methods which can be used for measuring D , τ , and R for release of constituents from monolithic materials. A laboratory protocol for estimating diffusion controlled release from compacted granular materials also has been developed [25]. In all cases, D is calculated based on rearrangement of Eq. (1) as

$$D = \frac{\pi M_t^2}{4t(\rho C_0)^2} \quad (3)$$

In the test method according to ANS 16.1, C_0 is equal to the total solid phase concentration of the product which results in smaller values of D and thus results in underestimation of the rate of constituent release. NEN 7435 and the granular diffusion test correct for this deficiency by defining C_0 as the experimentally determined availability.

The physical retardation factor is calculated based on use of sodium as the inert tracer as

$$\tau = \frac{D_{0,Na}}{D_{Na}} \quad (4)$$

The chemical retardation factor (R) for the component of interest is calculated from

$$R = \frac{D_{0,x}}{\tau D_x} \quad (5)$$

Table 3 presents diffusion coefficients and tortuosity values for several elements in untreated MWC residues and MWC residues incorporated into several solid products.

4. Estimation of constituent release for three management scenarios

Three broad management scenarios are provided for the purpose of illustration. Each of the categories are further divided into two specific applications:

Scenario 1. (Disposal in a percolation-controlled release regime)

Case A: unlined monofill with 20 cm infiltration per year;

Case B: unlined monofill with 200 cm infiltration per year.

Scenario 2. (Utilization as granular fill in a percolation-controlled release regime)

Case C: application of 0.5 m thickness with 20 cm infiltration per year;

Case D: application of 0.5 m thickness with 200 cm infiltration per year.

Scenario 3. (Utilization in road construction in a diffusion-controlled release regime)

Case E: compacted granular roadbase, wetted 35% of the time; and,

Case F: 50% substitution of natural aggregate in asphalt cement pavement, wetted 35% of the time.

This results in six possible cases to evaluate. Each of the six scenarios has been selected to illustrate reasonable field conditions but is not specific to any particular location. Table 4 provides a summary of the assumptions used for each disposal or utilization scenario and each MWC residue type. The disposal scenario is assumed to be an unlined scenario for illustrative purposes only because the approach involves estimation of cumulative release and adjacent soil impact. This is usually not the case for state-of-the-art disposal facilities which include liners and leachate collection. However, the cumulative release of a species still would provide useful information for the management of collected leachate. All of the cases are applied to bottom ash, combined ash and APC residues for illustration purposes. In some instances, notably bottom ash utilization in granular fill and road construction, the scenarios are examples of existing management strategies. In other instances, notably APC utilization in structural fill or road construction, the scenarios are not likely management strategies because of residue physical properties and chemical composition. They are included, however, to illustrate the approach and to provide considerations why particular strategies may be inappropriate. A sample set of results, using lead and chloride as examples, is provided for all 18 possible specific combinations. In two of the 18 combinations, bottom ash use as a compacted base course below an asphalt road and bottom ash use in asphalt pavement, field data from ongoing utilization studies are used to show that the approach does provide reasonable estimates of contaminant release. Release estimates from disposal of combined ash also are

Table 3

Typical ranges and values of tortuosity and diffusion coefficients for bottom ash, fly ash, APC residue, combined ash and several products incorporating MWC residues from mass burn MWC facilities. Chemical retardation factors can be calculated using Eq. (5)

$D_{0,x}$	Bottom ash [25]	APC residue [25]	Combined ash [25]	Grate ash in asphalt [32, 34]	Fly ash in asphalt [18]	Bottom ash in concrete paving blocks [18]
Tortuosity (—)	23	10	24	6600 ^a	8000	36
Diffusion coefficient (— $\log(m/s^2)$)						
Aa	9.04	na	na	na	12.6	11.1
Ba	9.7	na	12.1	na	14.7	3.9
Ca	9.1	11.1	12.8	13.5–14.2	15.2	14.1
Cd	9.14	> 15	> 15	na	15.8	> 14
Cl	8.69	10	10.5	11.6–12.2	11.4	12.2
Cr	9.23	12.3	10.4	na	> 14	> 13
Cu	9.11	> 14.7	14.6	na	14.4	13.6
K	8.71	10.1	10.2	na	12.6	10.4
Mg	9.04	> 14.7	15	14.2–15.2	na	na
Mn	9.1	na	na	na	na	na
Mo	9	na	na	na	14.4	12.8
Na	8.88	10.2	10.3	11.5–12.2	12.8	10.4
Pb	9.02	16.2	16.3	15.5–> 16 [35]	16.3	16.4
Zn	9.11	15.7	16	13.9–16.5	15.5	16.1

^a The values of tortuosity for grate ash in asphalt ranged between 440 and 14 200 with a mean value of 6600 for 8 measurements. Tortuosity was reported to be dependent on the extent of asphalt cracking, % asphalt cement and % bottom ash substitution for natural aggregate [35].

^b na = not available.

Table 4
Assumptions for estimating cumulative release for disposal and utilization scenarios

Disposal	Utilization as granular fill			Utilization as roadbase			Utilization as aggregate in asphalt ^a
	Case A	Case B	Case C	Case D	Case E	Case F	
Release interval (yr)	100	100	100	100	100	100	100
Mean temp. (°C)	20	20	20	20	20	20	20
Depth or thickness (m)	10	10	0.5	0.5	0.45	0.45	0.45
Infiltration (cm/yr)	20	200	20	200	na ^b	na	na
Time wet (%)	na	na	na	na	35	35	35
pD for Cl							
Bottom ash	na	na	na	na	10	13	13
APC residue	na	na	na	na	10	13	13
Combined ash	na	na	na	na	10	13	13
pD for Pb							
Bottom ash	na	na	na	na	15	16	16
APC residue	na	na	na	na	13	14	14
Combined ash	na	na	na	na	15	16	16
Bottom ash			APC residue		Combined ash		
Availability							
Cl (g/kg)	2.0		170		28		
Pb (mg/kg)	180		1000		500		
Leachate pH	8		11		6.5		
Pb Solubility (mg Pb/l)	0.04		10		0.1		
Cl Initial release (g Cl/l)	1.2		40		25		
Density (tonne/m ³)	2.1		1.4		1.9		

^a Assumes asphalt with 50% residue blended with 50% natural aggregate, 7 wt% asphalt cement.

^b Not applicable.

compared to field observations of leachate composition from monofill disposal. Verification with field data will obviously need to be expanded for many of the other management strategies to ensure that the approach has wide-spread applicability.

Estimation of constituent release for each of the above cases is specific to the range of anticipated field conditions and the element(s) or species of interest. The most critical field conditions are residue loadings (depth of residue fill, substitution rate, geometry, etc.), water contact, pH, oxidation or reduction potential, and temperature. The most critical MWC residue characteristics for developing field release estimates are constituent availability, solubility as a function of pH, the observed diffusion coefficient, and acid neutralization capacity. For all of the above scenarios, the compacted density and hydraulic conductivity will be similar for a given residue type. The primary difference amongst the scenarios is the amount of infiltration and the mode of water contact.

4.1. Scenario 1 – Disposal in a percolation-controlled regime

Disposal of MWC residues in a monofill results in very large mass of material located in a relatively limited area. Current monofill design and operating practices include the use of liners and leachate collection systems and the minimization of water infiltration through use of impermeable intermittent and final covers. During operation, residual moisture in the residues displaced by compaction and infiltration of precipitation may percolate through the material. Upon closure, infiltration is greatly reduced. This results in a low liquid to solid ratio ($LS < 1$) for long time intervals, e.g., decades or longer. Under this scenario, constituent release is controlled either by the solubility or availability of the species of interest.

Mean chloride concentrations observed from combined ash monofills during their first 5 years of operation ranged between 17 000 and 37 000 mg/l with maximum observed concentrations of 50 000 mg/l [19, 20]. If the availability of chloride in combined ash is approximately 35 000 mg/kg, from 50% to greater than 80% of the chloride present will be leached by the time an LS of 1 is achieved. Results from monitoring an ash monofill with bottom ash and fly ash over an interval of 20 yr indicated that chloride and TDS concentrations in leachate have begun to decline recently which corresponds to an LS of 0.6 [4].

Fig. 3 presents the pH and Eh of leachate from several MWC residue field lysimeters and monofills as a function of time. The observed pH of leachates from a bottom ash lysimeter and a monofill containing mixed bottom ash and fly ash, and combined ash monofills ranged from 8.5–10.5, 9.5–10.5, and 6.0–7.5, respectively. Note that reducing conditions are generated within the MWC residues relatively rapidly after disposal. Although bottom ash, APC residue and combined ash are all highly alkaline ($pH > 10$) immediately after generation, the pH of the leachates are less alkaline to nearly neutral for the cases of combined ash. This is most likely the result of carbon dioxide uptake from the atmosphere and carbon dioxide generation from microbial activity to form a carbonate buffer system. It is unclear whether the slightly acidic to neutral pHs observed for combined ash leachates are indicative of the environment within the monofill or reflect carbon dioxide uptake and oxidation

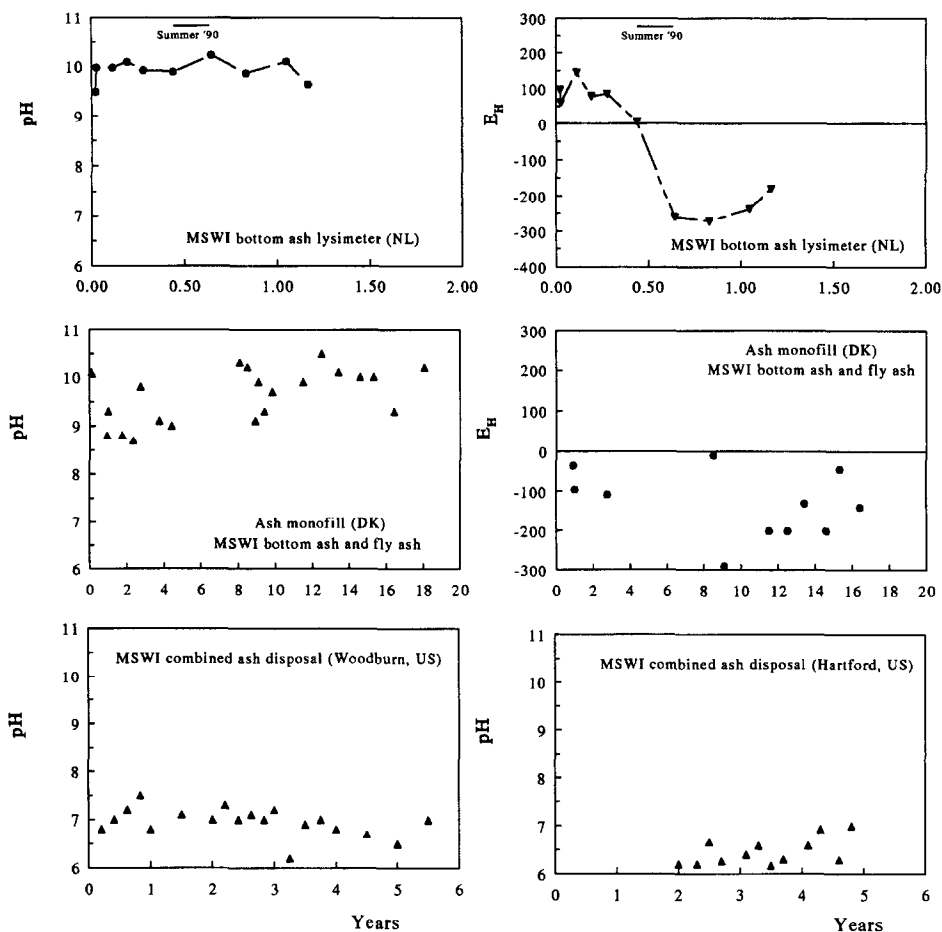


Fig. 3. pH and redox potential observed for monofill leachates (MSWI bottom ash [18]; Ash monofill (DK) [4]; Woodburn, combined ash [19]; Hartford, combined ash [20]).

which may have occurred during sample accumulation in leachate collection and storage systems. Oxidation of sulfide species to sulfuric acid or the production of greater concentrations of organic acids through biodegradation of uncombusted waste constituents may be responsible for the more acidic pHs observed for combined ash monofills. The mean alkalinity of leachates from three combined ash monofills ranged from 160 to 500 mg/l [20]. Thus, the initial conditions may be dictated by the characteristics of the residues, but external factors may result in considerable changes.

Fig. 4 presents observed lead concentrations in ash landfill leachate compared to laboratory testing results as a function of pH. In general, field observations are either similar to or less than laboratory concentrations measured at the same pH. Concurrently with the shift to a neutral pH, highly reducing conditions are established within the monofill. This is evidenced by observed elevated barium, iron and manganese

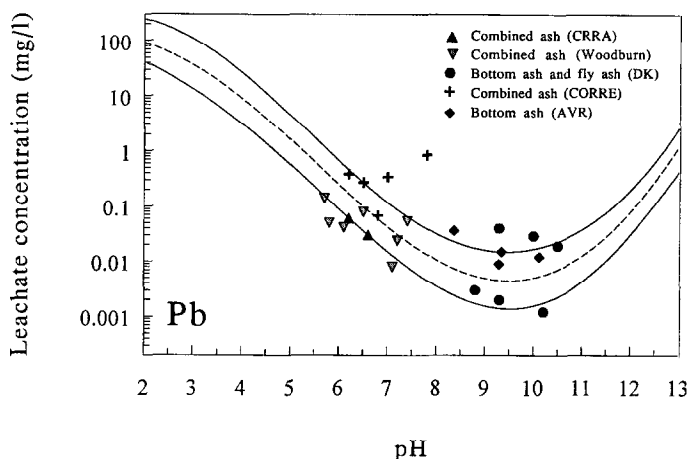


Fig. 4. Observed lead concentrations in ash monofill leachates compared to laboratory testing results as a function of pH (Combined ash (CRRA) [20]; Combined ash (Woodburn) [19]; Bottom ash and fly ash (DK) [4]; Combined ash (CORRE) [40]; Bottom ash (AVR) [39]). The dotted line indicates the log-mean concentration of laboratory results while the dashed lines indicate ± 1 standard deviation of log-transformed data.

concentrations in leachate [4, 20, 21]. These reducing conditions also result in significantly reduced heavy metal (e.g., lead, cadmium, etc.) concentrations compared to the oxidizing conditions usually present during laboratory testing. Caution should be exercised in obtaining field samples to avoid sample exposure to the atmosphere or oxidation.

The release of MWC residue constituents at a disposal site is primarily controlled by the solubility of those constituents. Several major constituents such as chloride, sodium, and potassium are highly soluble and their solubilities are independent of pH. The cumulative amount released will approach the availability for that species at low LS. The solubility of heavy metals is strongly dependent on the field pH and redox potential. The cumulative release of a specific constituent is the product of that constituent's solubility at field conditions and the LS.

The information required to estimate constituent release during disposal is the (i) disposal scenario geometry and physical conditions (e.g., type(s) of residue disposed, depth of residue fill, compacted residue density), (ii) anticipated infiltration rate, (iii) constituent availability, and (iv) constituent solubility as a function of pH for the residue types to be disposed. If the anticipated infiltration rate is not known, a LS of 2 is considered to be conservative for long term release from a monofill with a low permeability final cover. Highly soluble constituents (e.g., sodium, chloride, etc.) are considered to be released up to their availability. Sparingly soluble constituents are considered to be released at the concentration determined to correspond to the anticipated field pH. This estimate usually will be conservative because of decreases in solubility resulting from reducing conditions established in the field.

The anticipated LS is

$$LS = \frac{(\text{inf})(t_{\text{yr}})}{\rho H_{\text{fill}}} \quad (6)$$

and the estimated cumulative release is

$$M_x = (LS)(S_x), \quad (7)$$

where S_x is determined by solubility or availability, as appropriate.

Estimated releases of chloride and lead for two disposal cases for bottom ash, APC residue and combined ash are presented in Tables 5 and 6. Both Case A and Case B assume median values for availability for chloride and lead in each residue type, a monofill depth of 10 m and a release interval of 100 yr. Case A assumes an infiltration rate of 20 cm/yr ($0.02 \text{ m}^3/\text{m}^2 \text{ yr}$) while Case B assumes an infiltration rate of 200 cm/yr ($0.2 \text{ m}^3/\text{m}^2 \text{ yr}$). Case A assumes that infiltration is limited by monofill design while Case B may be more representative of a fill without aggressive infiltration control measures. Estimated release has been presented both on the basis of g Cl/kg ash disposed (or mg Pb/kg ash disposed), as calculated by Eq. (7) and on the basis of kg Cl/m² (or g Pb/m²). Release presented based on unit mass of residue disposed is useful for comparing the relative release or total environmental burden associated with different management scenarios while a release per unit area is useful for comparing the potential impact immediately underlying a disposal or utilization location. However, release would occur to the underlying soils only if the disposal sites were unlined. The actual impact underlying the disposal or utilization application frequently is reduced or minimized through use of liner systems and leachate collection. However, the release is not altered and the ultimate environmental burden is

Table 5
Estimated cumulative release of lead over 100 years for several management options

Management scenario	Bottom ash		APC residue		Combined ash	
	(mg/kg) ^a	(g/m ²) ^b	(mg/kg)	(g/m ²)	(mg/kg)	(g/m ²)
Disposal						
Case A	0.004	0.08	1	20	0.01	0.2
Case B	0.04	0.8	10	200	0.1	2.0
Utilization as granular fill						
Case C	0.08	0.08	30	20	0.2	0.2
Case D	0.8	0.8	300	200	2	2
Utilization as roadbase						
Case E	0.3	0.3	9	6	0.5	0.5
Utilization as aggregate in asphalt						
Case F	0.07	0.04	6	2	0.1	0.06

^a (mg/kg as disposed or utilized).

^b (g/m² land area impacted).

Table 6
Estimated cumulative release of chloride over 100 years for several management scenario

Management scenario	Bottom ash		APC residue		Combined ash	
	(g/kg) ^a	(kg/m ²) ^b	(g/kg)	(kg/m ²)	(g/kg)	(kg/m ²)
Disposal						
Case A	0.12	2.5	5.6	78	2.5	48
Case B	1.2	25	56	780	25	480
Utilization as granular fill						
Case C	2.0 ^c	2.1	110	77	28 ^c	27
Case D	2.0 ^c	2.1	170 ^c	120	28 ^c	27
Utilization as roadbase						
Case E	0.8	0.8	65	41	11	9.4
Utilization as aggregate in asphalt						
Case F	0.06	0.03	5	1.6	0.8	0.3

^a (g/kg as disposed or utilized).

^b (kg/m² land area impacted).

^c The cumulative amount of Cl⁻ released for this case is limited by the availability of Cl⁻ in the MWC residue (e.g., the maximum quantity has been released over the 100 yr interval).

a function of the leachate management scenario (e.g., treatment and discharge or alternative management practices). The release per unit area underlying an unlined disposal or utilization application is calculated as

$$M_t = \rho(H_{fill})(M_{\Sigma}). \quad (8)$$

The resulting estimates indicate that the release for Case B, as expected, is a factor of ten greater than for Case A for all residue types. Release of chloride from APC residue and combined ash is approximately 30 and 20 times greater, respectively, than that from bottom ash. Release of lead from APC residue and combined ash is approximately 200 and 2 times greater, respectively, than that from bottom ash. Furthermore, release of chloride typically is more than three orders of magnitude greater than release of lead for the same case and residue type.

4.2. Scenario 2 – Utilization as granular fill under a percolation regime

Utilization of MWC residues in a percolation controlled regime is also solubility controlled but differs from disposal in several critical aspects. The time period over which percolation occurs may be limited because the material may be exposed for a fixed period of time and then covered with a low permeability material or replaced. Typically the depth of fill material is limited and the material will remain oxidized during exposure. In addition, the properties of the other materials which may be in contact with the residues also can be important. Examples of utilization under a percolation regime are use of bottom ash or combined ash as a granular fill

material for embankments or as a gas venting layer in landfills. It is unlikely that APC residue would be used in this application because of inappropriate physical characteristics. This case is included here for comparative purposes only. Release under this scenario is extremely sensitive to the anticipated annual infiltration and cumulative LS.

The information required to estimate constituent release during utilization in a percolation regime is the (i) utilization scenario geometry and physical conditions (e.g., type(s) of residue utilized, depth of residue fill, compacted residue density, extent of gas exchange with the atmosphere), (ii) design life time of the residue use, (iii) anticipated infiltration rate, (iv) constituent availability, and (v) constituent solubility as a function of pH for the residue types to be utilized. Highly soluble constituents (e.g., Na, Cl, etc.) are considered to be released based on estimated initial release concentrations obtained from either laboratory column testing or prior field experience. Sparingly soluble constituents are considered to be released at the concentration determined to correspond to the anticipated field pH. However, the field pH and redox potential may be considerably different than the pH and redox potential of the residues measured under laboratory conditions. Uptake of carbon dioxide from microbial activity or atmospheric exchange can result in more neutral pH in the field than observed for recently generated residues tested under laboratory conditions. Reducing conditions often result in the field in the absence of atmospheric exchange either from microbial activity degrading residue organic carbon or from reduced metals within the residues. Care also must be taken to ensure that field samples are representative of pore water conditions and have not become oxidized or carbonated during sample accumulation (e.g., in leachate collection tanks) or sample handling.

LS values greater than 10 may be obtained over an interval of 100 years or longer for cases which have relatively high rates of infiltration. Under these conditions, several important changes in residue leaching chemistry may occur, including depletion of soluble species, changes in pH, etc. For this case, column leaching tests may provide a basis for determining the effects of depletion of solubility controlling species, e.g., chloride for cadmium, organic acids for copper, etc. Furthermore, column leaching tests can be carried out under oxidizing and reducing conditions to assess the effects of extreme redox potentials. Single extraction batch tests are not sufficient to determine these effects.

Release under a percolation-controlled regime is estimated first by calculating the anticipated LS using Eq. (6) and then calculating cumulative release using Eqs. (7) and (8). Estimated releases of chloride and lead from two percolation-controlled cases are presented in Tables 5 and 6 as Cases C and D. Both cases assume utilization of compacted residues at a fill depth of 0.5 m and a utilization period of 100 yr. Case C assumes infiltration of 20 cm/yr while Case D assumes infiltration of 200 cm/yr (analogous to Cases A and B). Calculation of chloride release indicates that the cumulative release for bottom ash and combined ash for both cases, and APC residue for Case D is equal to the availability. Approximately 65% of the available chloride is released for APC residue in Case C. While the release per unit mass of residue (g/kg) is significantly less for the disposal cases than the utilization cases with the same infiltration, the potential impact to the underlying soil (kg/m²) is similar for unlined

disposal and utilization with infiltration of 20 cm/yr. However for an infiltration of 200 cm/yr, the potential impact of the utilization case (Case D) is six to twenty times less than that estimated for the unlined disposal case (Case B) for the respective residue types because of limited chloride availability.

Estimated release of lead from combined ash is approximately twice the amount estimated for bottom ash for the same percolation controlled case. Estimated release from APC residue is more than three orders of magnitude greater than the release estimated for both bottom ash and combined ash. This result illustrates the effect of anticipated field pH on release. Estimated release of lead per unit mass of residue from the two percolation regimes is approximately 20 times greater than that estimated for unlined disposal cases at the same infiltration rate. This effect is from the greater LS achieved for the utilization scenarios. However, the potential impact to underlying soils is similar for both unlined disposal and utilization scenarios with similar infiltration rates because of solubility controlled release.

4.3. Scenario 3 – Utilization in road construction under a diffusion regime

The most frequently encountered scenario for release from utilized residues is the diffusion regime. For this scenario, residues are used as an aggregate substitute during the production of a product such as asphalt or Portland cement concrete, compacted in-place to a low permeability relative to surrounding materials, or compacted in-place and overlain by an impermeable material (e.g., in a roadbase with an asphalt surface layer). Flow of infiltrating water is around the residues or residue-containing products, and contact of these materials with the infiltration is limited to the boundary or external surface of the material (Fig. 5).

Estimation of constituent release for this scenario requires determination of the constituent availability, diffusion coefficient, mean field temperature, utilization time frame and the fraction of the time that the material interface is anticipated to be wetted. A 100 yr time frame is a very conservative estimate for the utilization of

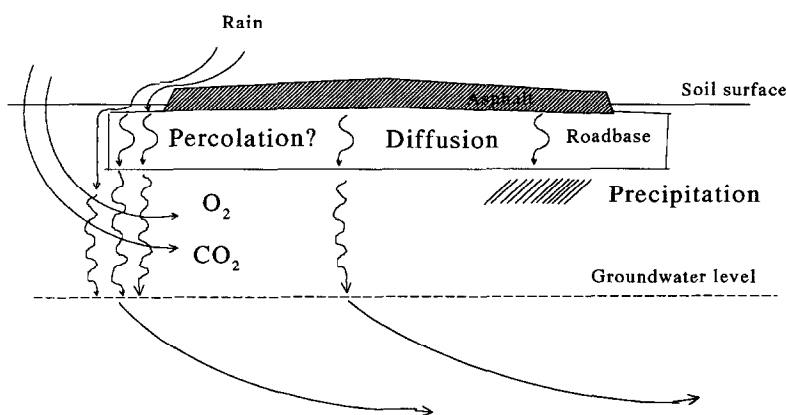


Fig. 5. Schematic diagram of percolation and diffusion controlled release conditions.

residues as an aggregate substitute in an asphalt pavement binder layer (the intermediate layer between granular base and surface layer), or as a compacted granular base underneath asphalt pavement. The fraction of the time that the material interface is wetted can be estimated based on the regional frequency of precipitation. A potentially more accurate approach to account for intermittent wetting would be to estimate the tortuosity based on the mean relative saturation and pore-size distribution of the compacted material [26]. However, estimation of mean relative saturation is not readily achieved from climatic data and future work is needed to evaluate the degree of saturation over time in the field.

The availability and diffusion coefficient leach parameters can be used to predict the release of contaminants during a given time period for a variety of application geometries. A 3-dimensional diffusion model enables consideration of application geometry and constituent depletion into account, so differences in leaching from a product with a cubic versus a flat rectangular shape can be described. With the 3-D model, release from only one side of the material also can be modeled.

A 3-dimensional model is based on the analytical solution of the linear diffusion from a parallelepiped, which initially is at a constant concentration, to an infinite region outside with a constant surface concentration [21, 27]. The diffusion profile is calculated in all three dimensions according to the equation:

$$\frac{C(x, y, z, t)}{C_0} = \frac{64(-1)^{l+m+n}}{n^3 l = 0 \quad m = 0 \quad n = 0 \quad (2l+1)(2m+1)(2n+1)} \cos \frac{(2l+1)\pi x}{2a} \\ \times \cos \frac{(2m+1)\pi y}{2b} \cos \frac{(2n+1)\pi z}{2c} \exp(-t\alpha_{l,m,n}), \quad (9a)$$

where

$$\alpha_{l,m,n} = \frac{\pi^2 D}{4} \left\{ \left(\frac{2l+1}{a} \right)^2 + \left(\frac{2m+1}{b} \right)^2 + \left(\frac{2n+1}{c} \right)^2 \right\}. \quad (9b)$$

Integration of the constituent flux across the surface boundary with respect to time results in an expression for calculation of cumulative release [21]:

$$M_t = \int_0^t \left\{ \left(\frac{64}{\pi^3} \right)^2 \frac{\rho C_0 abc}{l=0 \quad m=0 \quad n=0 \quad (2l+1)^2 (2m+1)^2 (2n+1)^2} \alpha e^{-t\alpha} \right\} dt. \quad (10)$$

Application of Eq. (10) permits estimation of the cumulative release of a constituent as a function of time. The cumulative release, expressed as fraction of the total leachable quantity (R_{\max}), can be calculated using the 3-D model for different product configurations and bulk applications based on the effective diffusion coefficient measured during laboratory testing or in the field under well-defined boundary conditions. The usual boundary condition applied for field translation is that the surface concentration of the leaching constituent is effectively zero.

The relative release from a roadbase (or slab exposed on one side) was calculated as a function of time for different effective diffusion coefficients ranging from $pD = 9$ to $pD = 12$ ($pD = -\log D$ with D in (m^2/s)). The cumulative release-time curves for this

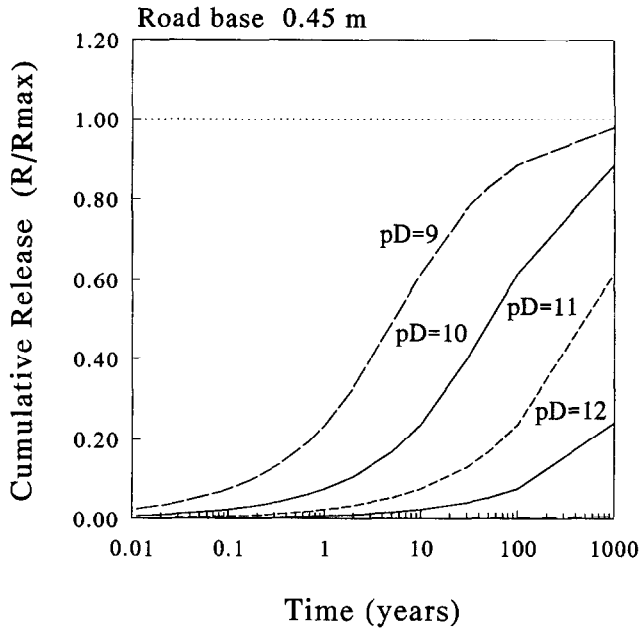


Fig. 6. Cumulative release from a roadbase (0.45 m thick) as a function of time for several values of pD estimated using the 3-dimensional diffusion controlled released model.

case, presented in Fig. 6, clearly indicate the significance of increasing pD (decreasing rates of diffusion). Greater than 60% of the available mass of a constituent with a pD of 9 will be released during the first 10 years, whereas less than 10% of the available mass of constituent with a pD of 12 will be released during a 100 yr interval.

Estimating release during utilization must consider adjustments to the pD measured in the laboratory to reflect anticipated field conditions. Temperature, the fraction of time the surface is wetted, and the degree of water saturation are important considerations. While the diffusion coefficient is a function of the diffusivity of the constituent of interest in water, tortuosity, and chemical retention, only diffusivity (D_0) is significantly a function of temperature. The temperature dependence of diffusivity in dilute ionic solutions can be considered to be proportional to the absolute temperature over limited temperature ranges [28], e.g.,

$$D_{T_2} = \frac{D_{T_1} T_2}{T_1} \quad (11)$$

The above relationship assumes that the viscosity of the pore water (leachate) does not change significantly over the temperature range of interest.

Alternatively, the effect of temperature on D has been correlated for release from cement stabilized products containing waste materials according to [29]:

$$pD_1 - pD_2 = 0.71 \left(\frac{1}{T_1} - \frac{1}{T_2} \right) \quad (12)$$

The cumulative release for a series of wet dry cycles can be approximated based on Eq. (1) by

$$\frac{M_t}{M_{t,w/d}} = \left(\frac{t}{t_{w/d}} \right)^{0.5} \quad (13)$$

or

$$M_{t,w/d} = M_t \left(\frac{t}{t_{w/d}} \right)^{-0.5} = M_t F_{w/d}. \quad (14)$$

Note that the calculation of M_t assumed a continuously water saturated material. Fig. 7 illustrates the effects of temperature and wet/dry cycles on diffusion controlled release from a 45 cm thick roadbase. The cumulative effect of these conditions can be significant over long time intervals.

Calculation of cumulative release curves for a variety of geometries and applications is impractical on a routine basis. The simple one-dimensional diffusion model presented in Eq. (1) can be used as an approximation. This is the same basis which was used to estimate the pD from tank leaching data. The initial and boundary conditions for this solution are (i) the initial constituent concentration is uniformly distributed in the matrix; (ii) the exposed surface for leaching has a liquid concentration which is maintained essentially at zero; and (iii) depletion does not occur. The advantage of this

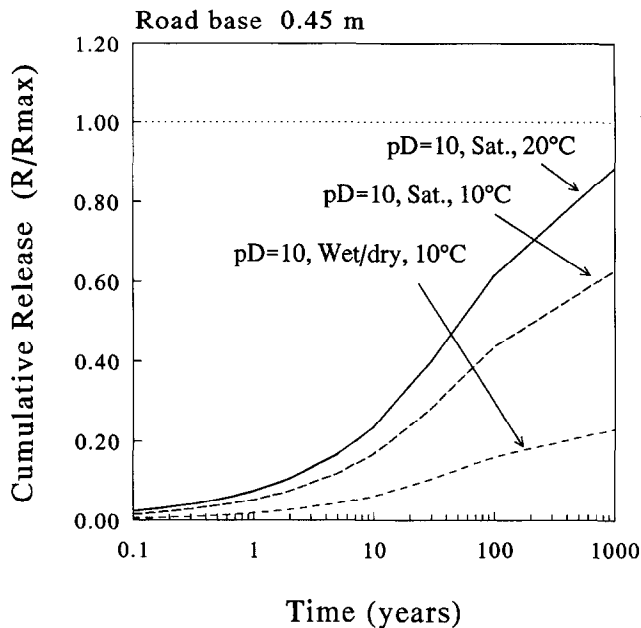


Fig. 7. The effects of temperature and intermittent wetting on cumulative release under diffusion controlled conditions.

approach is that cumulative release is only a function of the availability, diffusion coefficient and exposed geometric surface area. The one-dimensional model is independent of application-specific geometry. This approach is valid as long as the concentration in the material has not decreased substantially, avoiding species depletion. This approach assumes that the dimensions of the exposed surface area are large (e.g., this approach would be limited to short time intervals if applied to small blocks exposed on several faces). If depletion does occur over the time period of interest, the one-dimensional model will overpredict release, providing a conservative estimate for decision making. However, the amount of a specific element released will not exceed the availability of that element for all cases.

Fig. 8 provides a comparison of cumulative release estimated using the three-dimensional model and the one-dimensional model for a 45 cm thick roadbase (one surface exposed) for pD equal to 10 and 12. The effect of depletion on release is indicated by the significantly greater predicted release by the one-dimensional model for pD equal to 10. Differences between the model estimates are negligible for pD equal to or greater than 12 for a 100 yr release interval. However, in all cases the release estimated by the one-dimensional model will be equal to, or greater than the release predicted by the three-dimensional model. This confirms that the one-dimensional model provides a conservative estimate of release, provided that no significant changes in chemistry occur.

Interpretation of availability and pD information can be further simplified if a fixed time interval over which to consider release is defined. For example, a 100 yr interval

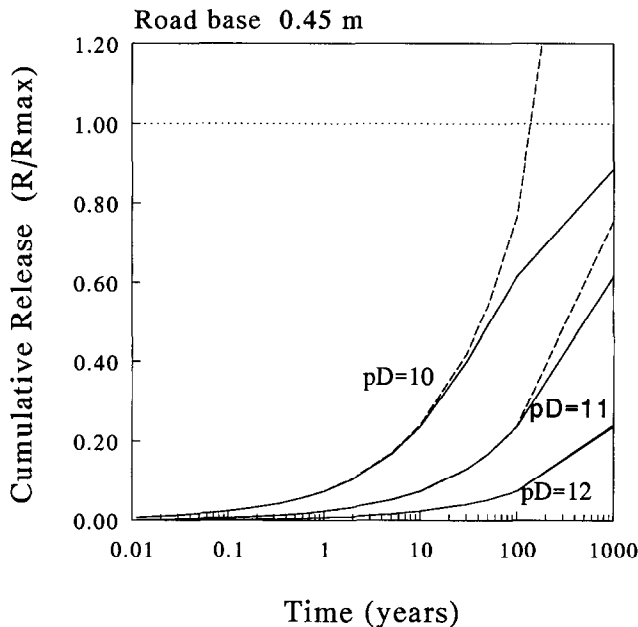


Fig. 8. A comparison of cumulative release estimates from 1- and 3-dimensional diffusion models.

may be considered a conservative estimate the useful life for roadbase or many other construction applications. Use of the one-dimensional model permits the development of charts which provide estimated cumulative release per unit area as a function of only availability and pD . These charts are referred to in this text as “cumulative release nomographs”. Examples of cumulative release nomographs are provided for lead and chloride in Figs. 9 and 10. Availability and pD are presented on the x - and y -axis, respectively. Diagonal lines indicate lines of constant cumulative release (e.g., 10, 100, 1000, etc. mg/m^2 over a 100 yr interval). Lines of constant cumulative release decrease in value from lower right to upper left of each figure. Data plotted above and to the left of a line of constant cumulative release provide less release than the indicated value; data below and to the right provide greater release than the indicated value.

Cumulative release nomographs provide a straightforward method of interpretation for evaluation of data from laboratory testing of diffusion-controlled release. An acceptable cumulative release can be defined for specific applications or locations based on evaluation of the potential impacts of that release. Data for applications

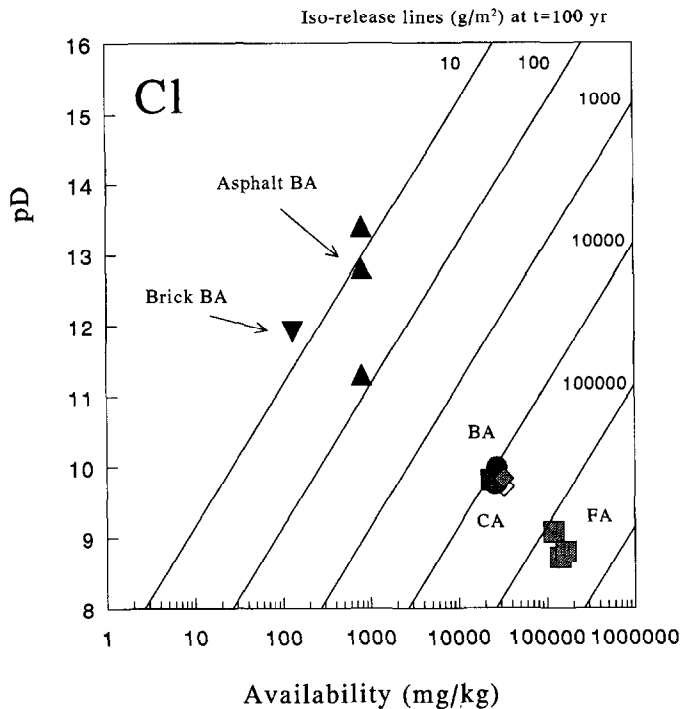


Fig. 9. A cumulative release nomograph for chloride release during a 100 yr interval. Typical values for untreated (open symbols) and treated (filled symbols) for bottom ash (BA, circles), APC residue (FA, squares) and combined ash (CA, diamonds) are presented. Treatment processes represented include bottom ash use in asphalt, bottom ash use in brick [18], and treatment of all three MWC residue types by four solidification/stabilization processes [25].

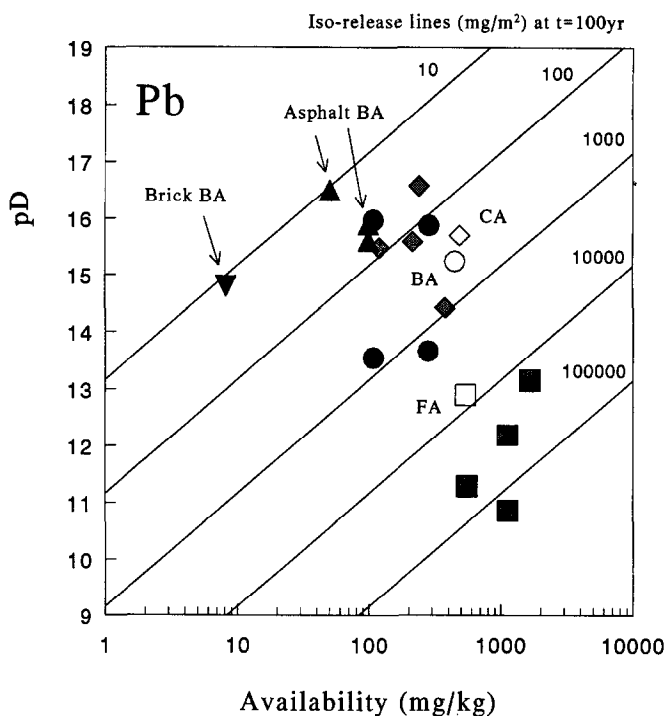


Fig. 10. A cumulative release nomograph for lead release during a 100 yr interval. Typical values for untreated (open symbols) and treated (filled symbols) for bottom ash (BA, circles), APC residue (FA, squares) and combined ash (CA, diamonds) are presented. Treatment processes represented include bottom ash use in asphalt, bottom ash use in brick [18], and treatment of all three MWC residue types by four solidification/stabilization processes [25].

which result in release less than the defined limit would be considered acceptable. Thus, diffusion-controlled release information can be readily incorporated into a regulatory framework.

The validity of this approach can be tested based on data obtained from two field studies. One field study has been the use of compacted bottom ash as a base course for construction of a service road in Rotterdam [30]. The roadbase is approximately 40 cm thick and is overlain by a clinker surface. The roadbase and underlying soils were cored after 10 yr of placement. Preliminary estimates of release of Pb and subsequent accumulation in the underlying soil indicated a cumulative release of between 0.1 and 0.4 g Pb/m² over the 10 yr interval. This compares favorably with the cumulative release estimated based on diffusion-controlled release of 0.38 g Pb/m² for a mean annual temperature of 10 °C and wetting 30% of the time. In addition, the released Pb was found to accumulate only in the uppermost few centimeters of soil underlying the pavement. Release of more mobile species such as Mo and sulfate were also in agreement with diffusion-controlled release from compacted granular

materials, but were found to have increased underlying soil concentrations to a depth of 10 cm. Field data were not available for chloride release. A statistically sound sampling program is in progress to obtain more reliable relations between laboratory predictions and field measurements [30].

In the second study, the release of ash constituents from asphalt pavements constructed with MWC bottom ash can be compared to estimates based on laboratory measurements of pD , R , τ , and availability for Cl and Pb. This study is one component of a program investigating the suitability of using MWC grate ash as an aggregate substitute in pavement applications [31–33].

The laboratory measurements of pD , R , t , and availability were carried out using the Dutch diffusion test (NEN 7345) and availability test (NEN 7341) [34]. Laboratory prepared pavement samples made with 25% to 75% bottom ash and 7% to 11% asphalt cement had pD 's of 11.3 to 12.8 for Cl. Availability was determined to be 1000 mg/kg and 100 mg/kg for Cl and Pb, respectively. While Cl can diffuse out of pavement monoliths made with MWC residues at low flux rates, Pb release is very sporadic, if at all and therefore a pD was not determined. Related studies to evaluate release from asphalt with varying degrees of laboratory imposed cracking, voids and aging have indicated pD values for Pb from 15.5 to greater than 16 [35].

The field evaluation of these types of materials involves a number of scenarios, including the construction of a binder course in a highway reconstruction application. One scenario involves the long-term leaching of exploratory pavement materials in large outdoor lysimeters. This scenario affords the opportunity to verify the diffusion modeling approach. The lysimeter contains 7246 kg of pavement fragments with approximate dimensions of 5 cm \times 30 cm \times 30 cm. The pavement was constructed with 2016 kg of dry <1.9 cm MWC bottom ash, 6048 kg of natural aggregate, and 10 wt% asphalt cement. This mix was compacted to approximately 2000 kg/m³ in a simple road segment, cured for 7 days, and excavated and placed in the lysimeter. The pavement fragments have been exposed to natural precipitation, dew, and some ponded infiltration in the lysimeter for over 3 yr. The measured pD for chloride release for this asphalt composition was 12 while the pD for lead could not be determined because the release was sporadic and not detected in the majority of laboratory extracts for this composition.

The analysis of Cl release is straightforward. During the three year period, approximately 170 g of Cl has been released. This accounts for 2.5% of the available Cl in the pavement ($R/R_{\max} = 0.025$). This cumulative release is in close agreement with one- and three dimensional modeling estimates for elements with pD of 12 (see Figs. 6 and 8). For the given time frame and estimated 35% exposure to wetted conditions, estimated exposed surface area (130 m²), and laboratory measured pD (12), the measured flux (470 mg/m²/yr) also is in close agreement with one-dimensional modeling estimates (500 mg/m²/yr). The analysis of Pb release is less straightforward. During the 3 yr period, the release of Pb has not been detected. Pb concentrations have always been below the detection limit of 0.005 mg/l.

Two examples can be used to illustrate estimation of constituent release under a diffusion-controlled regime over an extended time interval for comparison with the percolation-controlled release scenarios presented earlier. The first (Case E) is release

from a 45 cm thick road base constructed of compacted MWC residues (APC residues are included in this comparison although physical properties of the material would most likely prohibit their use in this application). The second example (Case F) is release from a 45 cm thick asphalt pavement base coarse layer which contained a 50 wt% substitution of natural aggregate with MWC residues and 7% voids. Case F assumes that a surface layer of asphalt material is overlain the base coarse and that the base coarse is in contact with the underlying soil. Thus, release is considered to be only from the bottom surface of the base coarse. The modeling of lead release from asphalt are conservative estimates for lead based on the intermittent and limited releases observed during laboratory testing of monoliths and field lysimeter data. Diffusion coefficients for release of Cl and Pb from combined ash and APC residues have been assumed to be the same values as those observed for release from bottom ash in asphalt. Availability for combined ash and APC residues in asphalt were assumed to be the same as for the untreated ash but reduced by the fraction of ash used. These are considered to be reasonable assumptions for initial estimates. Assuming a 100 yr utilization time interval, the cumulative release nomographs (Figs. 9 and 10) can be used to directly estimate release under saturated conditions. Approximately 1400 g Cl/m² and 0.5 g Pb/m² is estimated to be released during use of the compacted granular bottom ash while only approximately 50 g Cl/m² and 0.07 g Pb/m² would be released during use in the asphalt layer. Use of this material in a climate where it would be wetted only 35% of the time would result in reduction of these estimates by a factor of 0.59 (Eq. (13)). The resulting release for this condition is 800 g Cl/m² and 0.3 g Pb/m² for the compacted granular layer and 30 g Cl/m² and 0.04 g Pb/m² for the asphalt layer. This release also is equivalent to 800 mg Cl/kg ash and 0.3 mg Pb/kg ash for the compacted granular layer and 60 mg Cl/kg ash and 0.07 mg Pb/kg ash for the asphalt layer (Eq. (8)).

Release estimates for use of bottom ash, APC residue and combined ash both as a roadbase and as aggregate in asphalt are presented in Tables 5 and 6. Release of chloride from utilization of bottom ash in these applications is at least one order of magnitude less than release from use of combined ash or APC residue in a similar application. Release of lead from utilization of bottom ash is similar to that released from combined ash and at least one order of magnitude less than the release from use of APC residue in a similar application. Note that release during 100 yr interval is approximately 40% and 3% of the chloride availability for the compacted granular and asphalt uses of bottom ash, respectively, and less than 0.2% and 0.04% of the lead availability, respectively.

Estimated releases of chloride and lead from utilization of MWC residues in roadbase and asphalt applications can be compared to the disposal and utilization of these materials in a percolation regime. The potential impact of chloride to underlying soils from use of bottom ash in a roadbase application is less than half of that projected for disposal or utilization in a percolation regime with 20 cm/yr infiltration (Cases A and C). Use of these materials in an asphalt matrix reduces the potential impact from chloride release by one to three orders of magnitude compared to all other cases, depending on the cases which are compared. Release per unit mass of residue also is significantly less for all cases except when comparing APC residue

utilization in asphalt with disposal. The potential impact of lead from use of bottom ash and combined ash in an asphalt application is one-half to greater than one order of magnitude less than the other management options.

For all of the cases presented, if the lead estimated to be released is assumed to accumulate in the uppermost 10 cm of soil below disposal or utilization option, the resulting incremental increase in soil lead concentration would be 1 mg Pb/kg soil for every 0.1 g Pb/m² released. For example, utilization of bottom ash in the roadbase and asphalt pavement application described would result in a soil lead concentration increase of 3 and 0.4 mg Pb/kg soil, respectively. This is compared to guidelines that surficial soils should not exceed 85 mg/kg (The Netherlands) or 100 mg/kg (New Jersey, USA) in residential areas [36, 37]. In most cases, this incremental increase may not be distinguishable from natural variability and analytical errors.

5. Conclusions

Developing realistic estimates of constituent release from disposal and utilization of MWC residues requires a combination of laboratory tests which measure fundamental leaching parameters, mathematical modeling to carry out extrapolation of laboratory results to field scenarios, and field verification of critical assumptions. Single laboratory batch extractions, which form the basis of regulatory testing in many countries, do not provide sufficient information for predicting field behavior and may provide grossly misleading estimates (high and low) of potential environmental impact. Critical fundamental leaching parameters include availability, solubility as a function of pH, and diffusion coefficients for constituents of interest. Leaching behavior in the field is categorized as being controlled by either (i) constituent availability or solubility for percolation-dominated scenarios with loose granular residues, or (ii) controlled by diffusion for 'flow around' scenarios with compacted granular residues or monolithic products containing residues. Important field parameters include disposal or utilization application geometry, mode of liquid contact with residues or products (e.g., percolation or diffusion), infiltration rates and frequency, pH, temperature, and redox potential.

The approach presented here offers a different framework with which to evaluate residue management strategies. It departs significantly from the present practice of relying on single regulatory tests to predict leaching behavior. The approach appears promising in that for two, more complex scenarios (diffusion controlled release from compacted granular bottom ash used in base course below pavement and an asphalt road constructed with bottom ash as an aggregate substitute) the measured release of Pb from the compacted granular material and the release of Cl from the pavement agrees with modeled estimates.

Potential release of specific constituents which have low solubilities at or near neutral pH, such as lead, can be several orders of magnitude less than that which would be predicted by simple leaching tests. Conversely, highly soluble constituents, such as chloride, may be released up to their availability over relatively short intervals. Release of these constituents, while generally not regarded as toxic, may be of concern

in water catchment areas and areas of sensitive groundwater reserves (e.g., shallow aquifers with low flow conditions) because of potential contamination by total dissolved solids. This is especially a concern for management of APC residues and combined ash.

If the approach is valid, then the data presented in the 18 combinations of ash management scenarios can be evaluated as an illustrative exercise. For disposal scenarios, the leachates from APC residues will contain significantly more Cl and Pb than for combined ash or bottom ash, as the cumulative release of these constituents is higher. For unlined disposal situations, the cumulative soil impact is sensitive to the infiltration scheme. Nevertheless, APC residues will have a much more significant cumulative impact than bottom ash or combined ash. In all cases, bottom ash has the least significant cumulative impact. For utilization in granular fill applications, the leachates from APC residues will again contain significantly more Cl and Pb; the cumulative release and potential soil impact is highest with these residues as opposed to combined ash or bottom ash. The impact is again sensitive to the infiltration scheme. In all cases, bottom ash has the least significant cumulative impact.

For utilization in road construction, APC residues are again seen as having the most significant impact. It should be noted that scrubber residue use in structural fill or asphalt applications is also unlikely because of the physical characteristics of the material. It is believed that the incorporation of bottom ash into monolithic products (i.e., asphalt cement) or the placement of these materials below impermeable barriers such as an asphalt roadway results in diffusion-controlled release of contaminants from this material. The use of bottom ash as compacted base course below asphalt pavements or as aggregate substitute in asphalt pavements has a negligible cumulative release and cumulative impact.

Use of an approach which provides realistic release projections for a variety of management options from common laboratory data allows for comparison of advantages and disadvantages of specific disposal and utilization scenarios. It also permits focusing on which constituents may have significant environmental impact for further evaluation. For example, utilization of bottom ash as roadbase or incorporated in asphalt may be more environmentally protective than current disposal practices, depending on the specific conditions for each scenario.

The approach to estimating constituent release provided in this paper takes into consideration fundamental leaching parameters and disposal or utilization application scenario specific conditions. Further extensions of this approach should include consideration of external influences on release chemistry, such as externally moderated pH, redox potential and presence of complexing agents from other waste materials during co-disposal. Use of cumulative release nomographs allows for simplification of otherwise complex diffusion controlled release models so that they have potential for inclusion in a regulatory framework. Calculation of release per unit mass of MWC residue allows evaluation of potential overall environmental burden, while calculation of release per unit area of underlying soil allows evaluation of potential site specific impacts. It should be noted that this approach needs to be field validated for additional management scenarios, which requires longer term field data. This approach also may be extended to management of other waste materials.

Acknowledgements

The authors would like to acknowledge the International Ash Working Group (IAWG) which provided an extremely helpful forum for development of this approach. This work was supported in part by the United States Environmental Protection Agency (USEPA), Office of Research and Development, Risk Reduction Engineering Laboratory under Cooperative Agreement CR 818178. Although the information in this document has been funded in part by the USEPA, it does not necessarily reflect the view of the Agency and no official endorsement should be inferred.

Nomenclature

$C(x, y, z, t)$	concentration of a diffusing constituent within the solid matrix at location (x, y, z) and time (t) (mg/kg)
C_0	availability (mg/kg) = initial concentration (at $t = 0$) of the diffusing component in the solid matrix which must be uniformly distributed
D, D_x	diffusion coefficient for the component in the matrix of interest (m^2/s)
D_{Na}	diffusion coefficient of Na in the product (m^2/s)
$D_{0,x}$	diffusivity in water for component x (m^2/s)
$D_{0,Na}$	diffusion coefficient of Na in water ($pD_{Na} = 8.88$ at $22^\circ C$) (m^2/s)
D_{T_2}	diffusivity in water of constituent x at temperature T_2 (m^2/s)
D_{T_1}	diffusivity in water of constituent x at temperature T_1 (m^2/s)
$F_{w/d}$	fraction of the continuously wet cumulative release (M_t) which occurs, dimensionless
H_{fill}	height or depth of residue fill material (m)
Inf	annual infiltration (m^3/m^2 yr);
LS	liquid to solid ratio ($m^3/tonne$ or l/kg)
M_t	cumulative release per unit geometric surface area of product or fill (mg/m^2 or g/m^2)
M_Σ	cumulative release per unit mass of ash (mg x/kg ash or g x/kg ash)
$M_{t,w/d}$	cumulative release at time t for solid wetted only a fraction of the interval (mg/kg)
m_{site}	mass of residue disposed (tonnes)
R	chemical retardation factor of component x in the product, dimensionless
S_x	concentration of constituent x in aqueous solution at the anticipated field pH (mg x/l)
T_1	absolute temperature for which D_{T_1} is known (K)
T_2	absolute temperature for which D_{T_2} is desired (K)
t	time or elapsed time for release (s)
t_{yr}	time (yr)
$t_{w/d}$	time that the solid is wet during the interval (s)
a, b, c	parallelepiped dimensions (m)

erf()	standard error function
p()	– log()
x, y, z	location coordinates (m)
l, m, n	integers
ρ	density (kg/m ³)
τ	tortuosity or physical retardation in the product, dimensionless

References

- [1] A.J. Chandler, T.T. Eighmy, O. Hjelm, J. Hartlen, D.S. Kosson, H. van der Sloot and J. Vehlow, An international perspective on characterization and management of residues from municipal solid waste incineration, Summary Report of the IAWG, c/o Compass Environmental Inc., 2253 Belmont Ct., Burlington, Ontario, Canada, 1994.
- [2] D.S. Kosson, B.A. Clay, H.A. van der Sloot and T.T. Kosson, in: J.J.J.M. Goumans, H.A. van der Sloot and Th.G. Aalbers (Eds.), *Environmental Aspects of Construction with Waste Materials*, Elsevier, Amsterdam, 1994.
- [3] *Municipal Waste Combustion*, Proc. Internat. Specialty Conf. (Williamsburg, VA), Air and Waste Management Assoc., Pittsburgh, 1993.
- [4] O. Hjelm, *J. Hazard. Mater.*, this issue, 1995.
- [5] U.S. Environmental Protection Agency, Hazardous waste management system; Land disposal restrictions; final rule, Federal Register, Part II, Vol. 40 CFR Part 261 et seq. Washington, DC, November 1986.
- [6] A.M. Fallman, International Seminar on Leach Tests (Sept. 19, 1990), Swedish Geotechnical Institute, Linköping, pub. no. 317, 1990.
- [7] *Compendium of Waste Leaching Tests*, Report EPS 3/HA/7, Environment Canada, Ottawa, Canada, 1990.
- [8] A. Tessier, P.G.G. Campbell and M. Bisson, *Anal. Chem.*, 51 (1979) 844.
- [9] J.L. Frazer and K.R. Lum, *Environ. Sci. Technol.*, 17 (1983) 1.
- [10] C. Kheboian and C.F. Bauer, *Anal. Chem.*, 59 (1987) 1417.
- [11] P.M.V. Nirel and F.M.M. Morel, *Water Res.*, 8 (1990) 1055.
- [12] NEN 7341, Leaching characteristics of soil-, construction materials and wastes – Leaching tests – Determination of the availability of inorganic constituents for leaching from construction materials and waste materials, NNI (Dutch Standardization Institute, Delft), 1994.
- [13] State of California, Criteria for identification of hazardous and extremely hazardous wastes, California Code, Title 22, Article II: 1800.75-1800.82, 1985.
- [14] R.N.J. Comans, H.A. van der Sloot and P. Bonouvie, Proc. Municipal Waste Combustion. VIP 32. Air and Waste Management Association Pittsburgh, Pennsylvania. 1993, p. 667.
- [15] Test Methods for Solidified Waste Characterization, Acid Neutralization Capacity, Method #7, Environment Canada and Alberta Environmental Center, 1986.
- [16] R.C. Weast and M.J. Astle, *CRC Handbook of Chemistry and Physics*, 59th edn., CRC Press, West Palm Beach (FL), 1979.
- [17] NEN7343, Leaching characteristics of soil-, construction materials and wastes – Leaching tests – Determination of the leaching of inorganic constituents from granular construction materials and wastes, NNI (Dutch Standardization Institute, Delft), 1993.
- [18] C.W. Versluijs, I.H. Anthonissen and E.A. Valentijn, Mammoet 85. Report 738504008, National Institute for Public Health and Environment Publication, June 1990.
- [19] R.K. Cambotti and H.K. Roffman, Municipal Waste Combustion Ash and Leachate Characterization, Monofill – Fifth Year Study, Woodburn Monofill, Woodburn, Oregon, Report prepared by AWD Technologies, Pittsburgh, Pennsylvania, 1993.
- [20] D. Denfield, Personal communication: ash monofill leachate quality data, Connecticut Resources Recovery Authority, Hartford, Ct., 1994.

- [21] J. Crank, *The Mathematics of Diffusion*, 2nd edn., Oxford University Press, New York, 1989.
- [22] H.A. van der Sloot, G.J. de Groot and J. Wijkstra, in: P.L. Cote and T.M. Gilliam (Eds.), *Environmental Aspects of Stabilization and Solidification of Hazardous and Radioactive Wastes*, ASTM STP 1033, American Society for Testing and Materials, Philadelphia, 1989.
- [23] American Nuclear Society Standard Committee Working Group ANS 16.1, *American National Standard Measurements of the Leachability of Solidified Low-Level Radioactive Wastes by a Short-Term Procedure*, American Nuclear Society, La Grange Park, IL, 1986.
- [24] NEN 7345, *Leaching characteristics of soil-, construction materials and wastes – Leaching tests – Determination of the release of inorganic constituents from construction materials, monolithic wastes and stabilized wastes*, NNI (Dutch Standardization Institute, Delft), 1994.
- [25] D.S. Kosson, T.T. Kosson and H. van der Sloot, *Evaluation of Solidification/Stabilization Treatment Processes for Municipal Waste Combustion Residues*, NTIS PB93-229 870/AS, 1993.
- [26] C.E. Schaefer, R.R. Arands, H.A. van der Sloot and D.S. Kosson, *Prediction and experimental validation of liquid phase diffusion resistance in unstraturated soils*, *J. of Contaminant Hydrology*, in press.
- [27] G.J. de Groot and D. Hoede, *Verfijning van de beschrijving van de uitloging van reststofprodukten*, ECN-C-93-085. ECN publication, 1993 (3 D model)
- [28] R.C. Reid, J.M. Prausnitz and T.K. Sherwood, *The Properties of Gases and Liquids*, 3rd edn., McGraw-Hill, New York, 1977.
- [29] G.J. de Groot, H.A. van der Sloot, P. Bonouvrie and J. Wijkstra, *Karakterisering van het uitlooggedrag van intacte produkten*, Mammoet deelrapport 09, March 1990.
- [30] J.P.G.M. Schreurs, J. Peels, H.A. van der Sloot and L.G. Wesselink, manuscript in preparation.
- [31] I.E. Whitehead, T.T. Eighmy, D.L. Gress and X. Zhang, *Proc. Internat. Specialty Conference: An Environmental Evaluation of Bottom Ash Substitution in Pavement Materials*, Williamsburg, Virginia, 1993.
- [32] C.N. Musselman, M.P. Killeen, D. Crimi, S. Hassan, X. Zhang, D.L. Gress and T.T. Eighmy, in: J.J.J.M. Goumons, H.A. van der Sloot, Th.G. Albergs (Eds.), *Environmental Aspects of Construction With Waste Materials*, Elsevier, Amsterdam, 1994, p. 315.
- [33] C.N. Musselman, T.T. Eighmy, D.L. Gress, M.P. Killeen, J.R. Preshler and M.H. Sills, in: *Proc. 1994 National Waste Processing Conf.*, New York, New York, 1994, p. 83.
- [34] I.E. Whitehead, *An Environmental Evaluation of Bottom Ash Substitution in Pavement Materials*, Master Thesis, University of New Hampshire, Durham, New Hampshire, 1992.
- [35] T.T. Eighmy, D. Crimi, S. Hasan, X. Zhang and D.L. Gress, in: *Proc. of the 74th Transportation Research Board Meeting*, Washington, DC, Jan. 1995.
- [36] *New Jersey Interagency Task Force of the Prevention of Lead Poisoning, Sources of lead in the environment*, N.J. Department of Environmental Protection, 1994.
- [37] *Ministry of Housing Physical Planning and Environment Directorate-General for Environment Protection, Environmental Quality Standards for Soil and Water*, The Netherlands, 1991.
- [38] *Milieutechnisch onderzoek AVI slakken toepassing Rijksweg 15, Rijkswaterstaat, Dienst Weg en Waterbouwkunde*. Report D 0421-71-001, July 1992.
- [39] H.A. van der Sloot and D. Hoede, *AVI-bodemas als aanvulmateriaal*, ECN-c-91-044, 1991.
- [40] *US EPA (US Environmental Protection Agency), CORRE (Coalition on Resource Recovery and the Environment), Characterization of Municipal Waste and Combustion Ash, Ash Extracts, and Leachates*, EPA 530-SW-90-029A, March 1990.