

The stability of dolomite in landfill leachate-collection systems

Peter J. Bennett, Frederick J. Longstaffe, and R. Kerry Rowe

Abstract: This study uses several approaches to examine whether calcium-containing aggregate such as dolostone is a suitable drainage material for landfill leachate-collection systems. The thermodynamic stability of carbonate drainage materials has been assessed using published leachate data from landfills in the United Kingdom and leachate sampled from four large landfill sites of variable age in southern Ontario. Electron-microbeam techniques have been used to check for dissolution in dolomitic stone exhumed from the drainage layer of the Keele Valley Landfill leachate-collection system and from experiments that simulated landfill conditions. The mineralogy of cover soils applied daily to the landfill has been compared to the drainage stone and detrital material occluding pore space in the leachate-collection system to evaluate their relative contributions to clogging. The data suggest that dissolution of dolomitic drainage stone is not significant and contributes little to the clogging of landfill leachate-collection systems. However, crystallization of secondary calcite occurs about the dolomitic stones and sizeable quantities of inorganic fines, including dolomite, were present within some samples of "clog material" exhumed from the Keele Valley collection system. Most of the dolomitic fines probably were generated during construction of the collection system; such creation of fines ought to be minimized in future landfill developments.

Key words: leachate-collection system, landfills, clogging, mineralogy, leachate chemistry.

Résumé : Dans cette étude, on utilise plusieurs approches pour déterminer si des agrégats contenant du calcium tels que le dolostone constituent un matériel de drainage approprié pour les systèmes de captage des lixiviats de remblais sanitaires. La stabilité thermodynamique de matériaux de drainage constitués de carbonate a été évaluée en partant de données publiées sur les lixiviats de remblais sanitaires au Royaume Uni et de lixiviats échantillonnés sur quatre grands sites de remblais sanitaires de différents âges dans le sud-est de l'Ontario. Des techniques de micro-faisceaux d'électrons ont été utilisées pour vérifier s'il y a eu dissolution dans la pierre dolomitique prélevée dans la couche de drainage du système de captage de lixiviat du remblai de Keele Valley et dans les expériences qui ont simulé les conditions de remblai sanitaire. La minéralogie des sols de couverture qui ont été ajoutés quotidiennement au remblai a été comparée à la pierre de drainage et au matériel de débris qui colmatait l'espace interstitiel dans le système de captage de lixiviat pour évaluer leurs contributions relatives au colmatage. Les données semblent indiquer que la dissolution de la pierre dolomitique de drainage n'est pas significative et contribue peu au colmatage des systèmes de captage de lixiviat des remblais sanitaires. Cependant, la cristallisation de la calcite secondaire se produit autour des pierres dolomitiques, et des quantités assez importantes de particules fines inorganiques, incluant de la dolomite, étaient présentes à l'intérieur de certains échantillons du « matériau de colmatage » prélevés du système de captage de Keele Valley. La plupart des particules fines dolomitiques ont probablement été générées durant la construction du système de captage; une telle production de particules fines devrait être minimisée dans les futurs développements de remblais sanitaires.

Mots clés : système de captage de lixiviat, remblais sanitaires, colmatage, minéralogie, chimie des lixiviats.

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1. Introduction

Municipal solid waste (MSW) landfills should be designed to ensure a negligible impact on the present or future

use of groundwater beyond the site boundary (MOEE 1994). To minimize the likelihood of groundwater contamination, barrier systems are installed at the base of modern landfills. These barrier systems usually consist of a leachate-collection system overlying a natural or engineered liner of low hydraulic conductivity. The primary role of the leachate-collection system is to minimize the hydraulic gradient across the liner, which requires removal of leachate from the landfill (Rowe et al. 1995b).

Leachate underdrains typically consist of perforated pipes in a granular drainage layer beneath the waste. However, the effectiveness of the leachate-collection system can be degraded by the accumulation of organic and inorganic solids in the pore space of the drainage layer and within the perforated leachate-collection pipes (Brune et al. 1991; Rowe et

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al. 1995a, 1997; Fleming et al. 1999). This “clogging” can result in the retention of leachate within the landfill, increasing the hydraulic head across the liner and hence the potential for contamination of local groundwater.

An understanding of the processes responsible for clogging is essential in predicting the service life of landfill barriers and designing these systems for increased time to failure. Previous research has increased the general understanding of the clogging of leachate-collection systems (Brune et al. 1991; Rowe et al. 1995a, 1997; Bennett 1998; Fleming et al. 1999). However, it has not addressed adequately the possible contribution of the granular drainage stone itself to the formation of the clog, much of which is composed of calcite (Bennett 1998).

Brune et al. (1991) hypothesized that carbonate drainage stones were unsuitable for use in the drainage layer because they could dissolve and contribute to subsequent calcite crystallization on alkaline biofilms associated with the surface of drainage stone and collection pipes. This calcite would then occlude the pore space in the drainage layer, encrust the pipe perforation, and accumulate inside the pipe. Accordingly, the use of carbonate aggregate as a drainage material for landfill sites is discouraged by the United States Environmental Protection Agency (Daniel and Koerner 1993) and is forbidden in some states (Niemann and Hatheway 1997). However, Niemann and Hatheway (1997) concluded that typical landfill leachates would not dissolve significant amounts of carbonate aggregate. They passed leachate of pH 3 and 6–6.5 over carbonate aggregate in laboratory experiments and measured the change in mass of the drainage stones. Negligible weight loss occurred in the near-neutral pH range (6–6.5), leading them to conclude that significant dissolution of carbonate drainage stones would not occur in landfills. However, these findings were based solely on laboratory experiments that may not have been completely representative of field conditions.

Alternatives to carbonate aggregates in the leachate-collection system are not economically feasible in most heavily populated areas located in carbonate terrains, such as southern Ontario. Given the expense of alternatives, there is a surprising paucity of field evidence concerning the suitability of carbonate aggregate as drainage stone. In this paper, the putative contribution of dolomitic stone to clog formation is evaluated using geochemical and petrographic data for landfill leachate and drainage stone used in laboratory experiments and comparable materials exhumed directly from the Keele Valley Landfill (KVL) leachate-collection system. In particular, the questions of whether dissolution of the drainage stone contributes to the calcium burden of leachates and whether the physical migration of fines derived from the drainage stone comprises a significant fraction of the clog material are examined.

2. Methods

2.1. Field sites

Leachate was sampled from four southern Ontario MSW landfills. The Keele Valley Landfill (KVL) is located in the town of Maple, Ontario, 30 km north of Metropolitan Toronto. This 99 ha site has been accepting waste since the fall of 1983 and has a design capacity of 33 Mm³. The Brock-

West landfill (BWL), located in Pickering, Ontario, has been accepting waste since June 1975. It covers an area of 54 ha and contained approximately 18 Mt of waste at time of site closure in November 1996 (based on BWL filling records). The Beare Road landfill site (BRL), located in Scarborough, Ontario, has an areal extent of 81 ha and contained approximately 9.6 Mt of municipal and industrial waste by the time of site closure in 1983 (Gartner-Lee Limited 1990). The Halton Waste Management Facility (HL), located near Milton, Ontario, has been accepting waste since 1992, and in 1997 had an area of 10 ha (out of the eventual planned total of 53 ha of waste coverage by the time of site closure).

2.2. Leachate collection and analysis

At the BWL site, leachate was sampled from flooded landfill gas wells in recently filled areas (containing fresh waste and waste excavated from a smaller landfill site) and from a leachate-monitoring well in an older section of waste. These samples were obtained using a Solinst model 425 discrete interval sampler, which had been flushed and filled with helium prior to sampling. This procedure was intended to permit sample retrieval with minimal outgassing and atmospheric contamination. However, owing to technical difficulties, only one sample could be retrieved successfully using this technique (sample 1). For the other two BWL samples (samples 2 and 3), the discrete interval sampler was used as an open bailer. This approach may have allowed some outgassing of these samples. In addition, leachate samples were retrieved from main collection manholes at the KVL, BWL, HL, and BRL sites using an open bailer; these samples were also exposed to the atmosphere.

The temperature and pH of all samples were recorded at the time of collection using an Omega HH-25kc digital thermometer and an Oakton model 35624-30 portable pH meter (calibrated with pH 4, 7, and 10 buffer solutions). All samples except for sample 1 were stored in bottles provided by a commercial laboratory and submitted for analysis of standard water-quality parameters.

Sample 1 required special treatment because the volume of sample was limited to 0.125 L (the commercial laboratory required 1 L of sample), and precautions were necessary to ensure that the pressurized sample was not outgassed prior to analysis. This pressurized sample was split into two different containers: a 30 mL LDPE Nalgene sample bottle, which contained 200 μ L of reagent-grade concentrated HNO₃ (Caledon) for metal determination, and a 60 mL polypropylene syringe that contained HgCl₂ preservative for total dissolved inorganic carbon (DIC) analysis. Concentrations of some major and trace cations in leachate (Na, Ca, Fe, Mg, Mn, and Sr) were determined using a Perkin-Elmer Optima-3300 DV (system) inductively coupled plasma – atomic emission spectroscope (ICP–AES). An analytical blank containing 200 μ L of the same HNO₃ in deionized water was also analyzed. The DIC analysis of the pressurized sample in the syringe was performed using the method described by McAulliffe (1971).

2.3. Solids collection

Samples of dolomitic drainage stone and clog material were exhumed from 1 to 4 year old sections of the KVL leachate-collection system by researchers at the Geotechnical

Research Centre (GRC), the University of Western Ontario, London, Ont. The sampling locations and methods have been described previously (Rowe et al. 1995a; Fleming et al. 1999). These samples were obtained in 1993 and 1994 and had been air-dried and stored at room temperature since then.

Piles of unused drainage stone from the same quarry (dolomitic stone that had not been exposed to leachate or internal landfill processes) were also present at the surface of the landfill. These piles have been subjected to surface weathering for an unknown period of time. These stones, along with samples of daily cover soils, were collected in the summer of 1996.

Mesocosm experiments designed to simulate the environment of the leachate-collection system have been conducted at the GRC since June 1993 (Rowe et al. 1995a). The mesocosm design allows for the percolation of KVL leachate through a reactor consisting of a layer of refuse overlying a layer of crushed dolomitic stone (38 mm diameter). This stone originated from the same source used in the KVL collection system and is considered identical in provenance to the samples exhumed in the field. These experiments provided an opportunity to investigate the stability of dolomite in contact with landfill leachate. Upon termination of one mesocosm experiment in September 1997, drainage stone that had been exposed to the KVL leachate for over 4 years was collected and made into thin sections for analysis.

2.4. Textural, mineralogical, and petrographic analyses

The surfaces of the dolomitic samples were inspected for dissolution using a field-emission scanning electron microscope (Hitachi S-4500 SEM). In addition, thin sections of freeze-dried dolomite and clog material were prepared by a commercial laboratory (Core laboratories, Calgary, Alta.) using a process designed to preserve textures in semiconsolidated to unconsolidated materials. These thin sections were examined using a JEOL JXA 8600 Superprobe equipped with an energy-dispersive X-ray spectrometer (EDS) and back-scattered electron detector. The bulk mineralogy of the dolomitic drainage stone, clog material, and daily cover soils was determined using a Rigaku, high-brilliance, rotating anode, X-ray diffractometer (XRD) with a cobalt target.

3. Results and discussion

3.1. The stability of dolomite and other carbonates in landfills

3.1.1. Thermodynamic considerations

The stability of carbonate drainage stones in collection systems can be assessed on a thermodynamic basis using geochemical data for landfill leachate. Unfortunately, most available information has been obtained for leachate that travelled through the collection system and therefore had an opportunity to react with any drainage stone that might be present. However, Owen and Manning (1997) have reported geochemical results for leachates that had not yet reached the collection system. They collected samples from wells penetrating waste of variable age (less than 1 year to several decades) in three landfill sites servicing the United King-

dom. These data and the NETPATH code (Plummer et al. 1991) were used to calculate the saturation indices (SI) for the major calcium-containing minerals believed to contribute to the calcium load of leachate (Table 1). The thermodynamic stability of these minerals in contact with leachate can be assessed using the SI parameter, where a log SI value of greater than zero corresponds to supersaturation. All of the leachates sampled by Owen and Manning were supersaturated with respect to calcite, aragonite, and dolomite (among other carbonate minerals) and were undersaturated with respect to gypsum (and anhydrite). These data suggest that carbonate drainage stones would not dissolve in the presence of such leachates. However, because the leachates in the United Kingdom were collected using an open bailer, it cannot be ruled out that outgassing of dissolved CO₂ has caused their log SI values to be shifted to artificially high values. Resolution of this question would require samples for which in situ pCO₂ conditions were known more accurately.

The results of similar analyses for “virgin” (yet to pass through the collection system) leachate from the BWL site are listed in Table 2, along with the results for nonvirgin leachate samples from the BWL and three other southern Ontario landfill sites (KVL, HL, BRL) of varying age. Both virgin and nonvirgin leachates have log SI > 0 for calcium-containing carbonates, and log SI < 0 for gypsum. Similar to the examples from the United Kingdom, it seems that dolomite and other carbonates are thermodynamically stable in southern Ontario landfills, whereas gypsum is not. Nevertheless, it is acknowledged that the lowest (but still positive) log SI values, including that for dolomite, were obtained for sample 1, the one sample for which outgassing of leachate was prevented. Of all the data, the pCO₂ conditions for this sample are most representative of the depth from which it was retrieved.

It is useful to compare the log SI values for the southern Ontario landfills in the context of their variability in drainage-layer types. The BRL site has no engineered system for the control of leachate, other than collection by pumping from the southwest corner of the site (Desrocher and Sherwood-Lollar 1997). Its leachate has the lowest (but still positive) log SI values for carbonate, including dolomite (Table 2, sample 4, 0.179), of all samples that represent average landfill effluent (Table 2, samples 4–7). BWL does not have a drainage layer, only perforated pipes surrounded by a small amount of pea gravel of unknown composition. Leachate from KVL has passed in part through a dolomitic stone drainage layer and in part through a widely spread French drain system (a pipe surrounded by a small amount of crushed dolomitic stone). Leachate from HL, which is draining the youngest waste of all four landfills, has travelled through a layer of crushed dolomitic stone (Rowe 1998). Despite these differences in potential for contact with dolomitic drainage stone (or the age of waste drained), the log SI values are very similar for leachate from the main collection manholes at BWL, HL, and KVL (Table 2, samples 5–7). Data for the BWL site also illustrate that log SI values for carbonate minerals, dolomite in particular, increase substantially from higher in the waste (Table 2, samples 1 and 2, 0.004 and 0.957) to just above the collection system (Table 2, sample 3, 2.900). However, the increase in log SI is very modest from that point in the landfill to the main

Table 1. Saturation indices for leachates sampled at sites in the United Kingdom in 1993 and 1994 (data from Owen and Manning 1997).

| Well No. | Log SI _{calcite} | Log SI _{aragonite} | Log SI _{dolomite} | Log SI _{gypsum} |
|------------------------|---------------------------|-----------------------------|----------------------------|--------------------------|
| Arpley 1993 | | | | |
| A1 | 1.564 | 1.415 | 3.069 | -2.990 |
| A2 | 1.304 | 1.151 | 3.116 | -1.994 |
| A3 | 1.454 | 1.309 | 3.353 | -2.363 |
| A4, A7 | 1.380 | 1.229 | 2.614 | -2.333 |
| A5 | 1.850 | 1.708 | 3.916 | -1.810 |
| A6 | 1.247 | 1.093 | 2.502 | -2.713 |
| A8 | 0.510 | 0.362 | 2.083 | -1.295 |
| A9 | 1.325 | 1.180 | 3.137 | -2.607 |
| Arpley 1994 | | | | |
| A1 | 1.887 | 1.738 | 4.264 | -3.103 |
| A2 | 1.732 | 1.580 | 4.157 | -4.131 |
| A3 | 2.073 | 1.930 | 4.185 | -3.129 |
| A4, A7, A9 | 1.688 | 1.540 | 3.937 | -3.914 |
| A5 | 1.628 | 1.485 | 4.279 | -3.670 |
| A6 | 1.691 | 1.537 | 3.282 | -1.602 |
| A8 | 2.048 | 1.902 | 4.190 | -2.690 |
| A10 | 2.266 | 2.125 | 3.817 | — |
| Danes Moss 1993 | | | | |
| DM1 | 0.310 | 0.153 | 0.173 | -1.244 |
| Danes Moss 1994 | | | | |
| DM1 | 2.186 | 2.034 | 4.435 | -2.691 |
| Maw Green 1993 | | | | |
| MG1 | 0.933 | 0.788 | 2.143 | -2.188 |
| MG2 | 0.954 | 0.802 | 2.592 | -3.275 |
| MG3 | 0.489 | 0.334 | 0.832 | -2.065 |
| MG4 | 0.273 | 0.118 | 0.300 | -1.118 |
| MG5 | 0.954 | 0.803 | 2.452 | -1.688 |
| MG6 | 0.897 | 0.758 | 2.431 | -3.060 |
| MG7 | 0.700 | 0.556 | 1.854 | -1.830 |
| MG8 | 0.778 | 0.633 | 1.935 | -2.763 |
| MG9 | 0.805 | 0.662 | 2.664 | -3.529 |
| MG12 | 0.663 | 0.519 | 1.385 | -1.921 |
| MG13 | 2.157 | 2.009 | 4.209 | -2.209 |
| MG14 | 1.217 | 1.071 | 2.801 | -2.247 |
| Maw Green 1994 | | | | |
| MG1 | 1.067 | 0.924 | 2.262 | -2.475 |
| MG2 | 1.414 | 1.264 | 3.384 | -3.378 |
| MG3 | 1.100 | 0.947 | 2.204 | -1.896 |
| MG4 | 0.948 | 0.792 | 1.717 | -1.671 |
| MG5 | 1.175 | 1.024 | 2.927 | -3.238 |
| MG6 | 1.374 | 1.237 | 3.422 | -2.764 |
| MG7 | 1.221 | 1.082 | 3.168 | -2.280 |
| MG8 | 1.302 | 1.163 | 3.022 | -2.429 |
| MG9 | 1.565 | 1.422 | 3.485 | -1.881 |
| MG12 | 1.099 | 0.957 | 2.335 | -2.177 |
| MG13 | 1.551 | 1.411 | 0.870 | -3.024 |
| MG14 | 2.006 | 1.867 | 3.996 | -0.583 |

manhole, where leachate exits the collection system (Table 2, sample 5, 3.056).

To summarize, the possibility that dolomite is not thermodynamically stable in some areas of the landfill cannot be ruled out completely. Leachate samples that are more representative of in situ landfill conditions (i.e., not exposed to the atmosphere) are required to confirm such a conclusion. In addition, geochemical calculations can be sensitive to the

geochemical modelling procedure used; similar trends but different absolute log SI values can be obtained, depending on the model that is used. That being said, the samples analyzed in this study did not have geochemical characteristics that would promote dissolution of carbonate drainage stone.

The geochemical evolution from fresh water that infiltrates landfills to carbonate-supersaturated leachate that exits from their base involves the addition of Ca²⁺, Mg²⁺, and

Table 2. Leachate chemistry and saturation indices for southern Ontario landfills.

| Sample No.: | 1 | 2 | 3 | 4 | 5 | 6 | 7 |
|-------------------------------------------------------------------|-----------|-----------|-----------|------------------|---------------------|--------------------|---------------------|
| Site: | BWL-FGW | BWL-FGW | BWL-MW | BRL ^a | BWL-MM ^a | HL-MM ^a | KVL-MM ^a |
| Date | 21-8-1997 | 25-6-1998 | 25-6-1998 | 6-10-1997 | 25-6-1998 | 6-10-1997 | 5-8-1997 |
| Temp. (°C) | 15.0 | 17.5 | 35.5 | 13.0 | 35.0 | 13.0 | 20.0 |
| pH | 6.10 | 6.40 | 7.50 | 6.83 | 7.47 | 7.60 | 6.93 |
| Sodium (Na; mg/L) | 836 | 875 | 2 600 | 622 | 3 220 | 662 | 2 560 |
| Potassium (K; mg/L) | — | 127 | 1 220 | 118 | 892 | 316 | 1 200 |
| Calcium (Ca; mg/L) | 1 585 | 447 | 32.8 | 280 | 110 | 205 | 867 |
| Magnesium (Mg; mg/L) | 377 | 275 | 91.6 | 68.1 | 142 | 294 | 435 |
| Bicarbonate (as HCO ₃ ⁻ ; mg/L) | 2 221 | 2 400 | 9 880 | 1 190 | 4 380 | 3 270 | 8 990 |
| Sulphate (SO ₄ ²⁻ ; mg/L) | — | 12 | 114 | 33 | 89 | 81 | 189 |
| Chloride (Cl ⁻ ; mg/L) | — | 1 780 | 4 830 | 1 220 | 7 450 | 681 | 3 770 |
| Reactive silica as SiO ₂ (mg/L) | — | 30.6 | 50.4 | 16.7 | 38.3 | 24.6 | 50.3 |
| Nitrate and nitrite (mg/L NO ₃ , NO ₂ as N) | — | 0.22 | 0.61 | 0.08 | 0.22 | 0.45 | <0.05 |
| Phosphorus (dissolved ortho as P; mg/L) | — | 0.01 | 3.77 | <0.01 | 2.14 | 0.90 | <0.01 |
| Ammonia (as N; mg/L) | — | 136 | 1 630 | 113 | 750 | 228 | 20 |
| Conductivity (µS) | — | 7 580 | 26 300 | 4 420 | 24 700 | 6 360 | 22 000 |
| Dissolved organic carbon (mg/L C) | — | 172 | 1 520 | 244 | 1 710 | 199 | 5 300 |
| Iron (Fe; mg/L) | 563 | 66.4 | 15.2 | 658 | 1.90 | 4.96 | 166 |
| Manganese (Mn; mg/L) | 377 | 3.86 | <0.2 | 0.82 | 0.22 | 0.40 | 6.81 |
| Log SI _{calcite} | 0.200 | 0.454 | 0.972 | 0.305 | 1.227 | 1.389 | 1.61 |
| Log SI _{aragonite} | 0.049 | 0.304 | 0.836 | 0.152 | 1.090 | 1.236 | 1.463 |
| Log SI _{dolomite} | 0.004 | 0.957 | 2.900 | 0.179 | 3.056 | 3.116 | 3.231 |
| Log SI _{gypsum} | — | -2.404 | -3.020 | -2.012 | -2.439 | -1.855 | -1.259 |

Note: Sample 1, flooded gas well (FGW), sampled from 12 m below grade with leachate at 7.3 m below grade, and collected with minimum outgassing and atmospheric contamination; sample 2, FGW, collected from 9.1 m below grade; sample 3, monitoring well (MW), collected just above leachate-collection system; sample 4, no leachate-collection system, pumped from northwest corner of site; samples 5–7, collected from main manhole (MM), samples effluent from leachate-collection system. —, no data.

^aLeachate had already passed through the collection system.

HCO₃⁻ during chemical, physical, and microbial processes. Bicarbonate can be derived from dissolution of cover-soil carbonates or other carbonates present in the waste, assuming log SI < 0 during the earlier stages of chemical evolution towards a leachate. However, it originates mostly from the mineralization of organic carbon by microbes such as methanogens (Brune et al. 1991; Owen and Manning 1997; Bennett 1998). Potential sources for Ca²⁺ (or Mg²⁺) in the waste are not as well documented. Dissolution of calcite and dolomite present in the waste and cover soils are viable sources of calcium and magnesium early in leachate evolution. However, the data for the leachates from the United Kingdom (Table 1) strongly suggest that carbonate saturation is reached very early (<1 year) in their evolution. Leachates can acquire additional calcium in other ways, for example, through the degradation of organic matter (calcium can exceed 1% dry weight in some plant tissues; Salisbury and Ross 1992). The disposal of construction waste, especially wallboard, is another potentially important source for calcium and may be particularly significant given the thermodynamic instability of gypsum in landfill leachates.

3.1.2. Petrographic observations

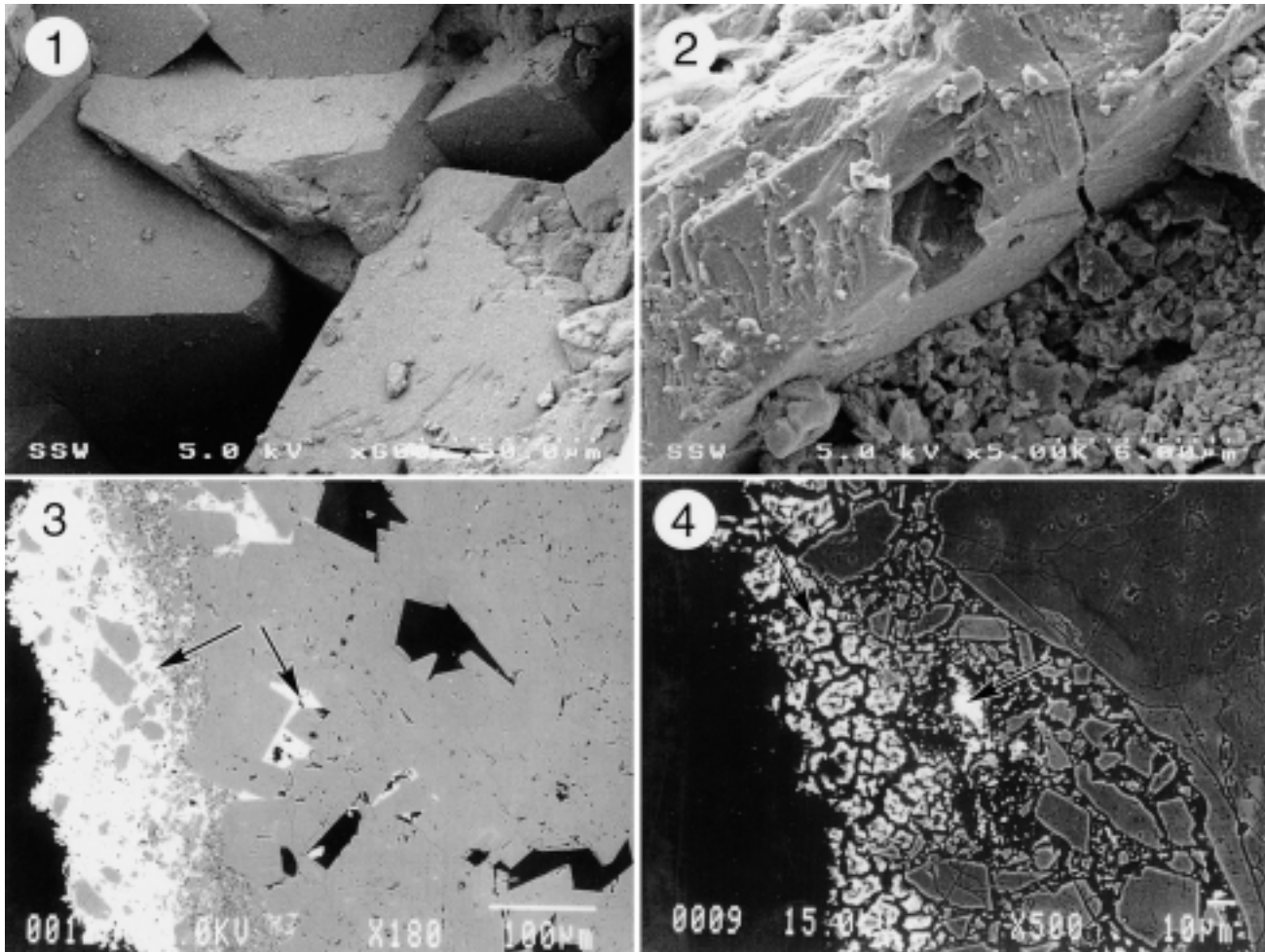
In the very early stages of leachate evolution, when acetogenesis is dominant within the landfill and pH is low, conditions may be such that carbonate undersaturation is prevalent. In this hypothetical scenario, dolomitic drainage stone may begin to dissolve in contact with potentially acidic leachate until saturation with respect to dolomite is

reached. This process should be detectable on the surface of dolomitic stone from a leachate-collection system. Such samples were obtained during the exhumation of a 2 year old section of the KVL collection system. These stones were covered in a thick, viscous biofilm (Rowe et al. 1995a), which was removed so that the inorganic surface of the stone could be examined. The surface of dolomitic stone taken from a pile on the landfill site was also examined for signs of dissolution.

Incipient dolomite dissolution is commonly characterized by the formation of small pits, which can be detected at high magnification on crystal surfaces (Herman and White 1985). The surface of the dolomitic stone exhumed from the KVL leachate-collection system is devoid of such pits (Fig. 1), whereas similar stone taken from the pile contains abundant small holes on its surface (Fig. 2). The latter features may reflect dissolution during acid rainfall. Surface features comparable to those of the pit shown in Fig. 2 were not observed in any samples exhumed from the KVL leachate-collection system, although microporosity inherited from the parent dolomite remains in the interior of some aggregate samples.

Although the leachate may be generally supersaturated with respect to dolomite, it is possible that dissolution has occurred at locations where bacteria are in contact with the drainage stone (membrane effects) and (or) prior to the establishment of a biofilm, which generally has a pH of 8. Back-scattered electron images (BSE) of dolomite from the mesocosm experiments, which were designed to emulate the environment of a landfill leachate-collection system, are

Fig. 1. SEM image of the surface of dolomitic drainage stone exhumed from the KVL leachate-collection system. No pits or other irregularities that might indicate dissolution by leachate were observed in this or other samples. Loose debris on the surfaces is likely an artifact of sample preparation. **Fig. 2.** SEM image of the surface of dolomitic drainage stone exposed to surface weathering while stored in an aboveground pile at the landfill. Irregularities in the surface texture of the stone are present, including a pit in the dolomite crystal (centre of the image). These features suggest incipient dissolution of the stone, presumably as a consequence of local acid rain. Loose debris on the surfaces is likely an artifact of sample preparation. **Fig. 3.** BSE image of dolomitic drainage stone from a laboratory mesocosm. Arrows show secondary calcite cement (white) that crystallized on the surface of the drainage stone (grey) over 4 years. The boundary between the stone and the overlying secondary calcite plus entrained detrital dust fragments (mostly dolomite) is very sharp. This suggests that dolomite dissolution during the experiment was very limited, or did not occur. Some original porosity near the surface of the dolomitic drainage stone has been filled by secondary calcite, and indicates leachate penetration to this depth. Primary porosity farther within the interior of the stone (black areas) is devoid of this calcite. **Fig. 4.** BSE image of dolomitic drainage stone exhumed from a laboratory mesocosm. Arrows point to secondary Fe sulphide that formed on the outer edge of the detrital dust fragments that mantle the stone. The boundary with the stone has remained very sharp and shows no obvious dissolution features. The very thin white lines around the stone and fragment edges are instrumental artifacts.



useful in assessing the potential significance of such membrane effects (Figs. 3, 4). The samples of dolomitic stone that were studied had been exposed to leachate with a pH ranging from 6.2 to 8.0 for over 4 years. The coating of calcite around the stone in Fig. 3 crystallized during the mesocosm experiment. This calcite has also filled primary porosity within the stone to about 100 μm below its surface, indicating that leachate penetrated to such a depth.

It is not obvious whether the calcite cement formed directly as an inorganic precipitate from leachate or as the result of microbially induced crystallization associated with the biofilm on the surface of the drainage stone. Regardless,

the sharp grain boundaries between the stone and secondary calcite cement suggest that little dolomite dissolution occurred prior to or during formation of calcite. Fragments of dolomite are entrained in the calcite cement that coats the stone. This “dust” probably was generated mechanically, likely when the stone was initially placed in the site and spread by bulldozers. The dolomite fragments were then transported by the leachate until they became cemented by secondary calcite.

Figure 4 illustrates a different region of the same drainage stone shown in Fig. 3. The white material (arrows) located around the outer portion of the dust envelope is secondary

Table 3. Bulk mineralogy of cover soils and solid “clog” material from the Keele Valley Landfill.

| Sample No. | Calcite | Dolomite | Quartz | Feldspars | Dolomite / (quartz + feldspars) |
|--------------------------|---------|----------|--------|-----------|---------------------------------|
| Daily cover soils | | | | | |
| Old cover soil 1 | 11 | 11 | 65 | 12 | 0.14 |
| Old cover soil 2 | 15 | 0 | 71 | 14 | 0 |
| New cover soil 1 | 36 | 7 | 42 | 15 | 0.12 |
| New cover soil 2 | 36 | 12 | 39 | 13 | 0.23 |
| Clog | | | | | |
| Sample 1 | 71 | 14 | 11 | 4 | 0.93 |
| Sample 2 | 75 | 0 | 17 | 8 | 0 |
| Sample 3 | 75 | 0 | 19 | 6 | 0 |
| Sample 4 | 21 | 9 | 63 | 7 | 0.13 |
| Sample 5 | 83 | 4 | 13 | 0 | 0.31 |
| Sample 6 | 83 | 3 | 10 | 4 | 0.21 |
| Sample 7 | 67 | 7 | 22 | 4 | 0.27 |
| Sample 8 | 18 | 22 | 52 | 9 | 0.36 |
| Sample 9 | 92 | 0 | 7 | 0 | 0 |
| Sample 10 | 88 | 3 | 8 | 2 | 0.30 |
| Sample 11 | 36 | 29 | 31 | 3 | 0.85 |
| Sample 12 | 62 | 18 | 18 | 2 | 0.90 |

Note: Amounts were estimated from semiquantitative XRD analysis using peak heights of the most intense diffraction for each phase.

Fe sulphide. There is a difference between the distribution of secondary Fe sulphide (Fig. 4) and calcite (Fig. 3). Unlike the calcite, the Fe sulphide does not penetrate the surface of the stone. Its localized distribution on the outer surface of the dust envelope suggests that secondary sulphide formation is related to the biofilm that coated this area. Localized distribution of microbial communities in both the KVL leachate-collection system and the mesocosms, including the sulphate reducers that helped to form the Fe sulphide, have been reported previously (Rowe et al. 1995a; Fleming et al. 1999). In summary, despite the abundant presence of microbes, the boundary with the dolomitic drainage stone has remained very sharp. There is little evidence that microbial membrane effects caused significant dissolution of the dolomite.

3.2. The contribution of dolomite fines to inorganic clog formation

The presence of fines in the leachate could enhance the rate of calcite crystallization in the leachate-collection system by providing abundant sites for heterogeneous nucleation. Accordingly, we have examined whether dolomitic drainage stone has contributed unduly to this process.

The bulk mineralogy of KVL daily cover soils and exhumed inorganic clog material is summarized in Table 3. Daily cover soils can be rich in calcite and dolomite, although there is a large variation among the four samples analyzed. The inorganic fraction of the clog samples is composed predominantly of calcite, but also contains detrital minerals that likely originated from cover soils, overlying waste, and the dolomitic drainage stone itself. Quartz, K-feldspar, and plagioclase are common cover-soil minerals and were detected in varying quantities in the inorganic clog matrix. These minerals were likely entrained in the leachate and carried into the collection system from the overlying waste. Fleming et al. (1999) also noted that minerals originating from cover soils were present in the clog material and

attributed this feature to the absence of a geotextile filter between the base of the waste and the drainage blanket of the leachate-collection system.

Dolomite is also present in many of the clog samples. There are two possible sources for dolomite in the KVL leachate-collection system: (1) daily cover soils, and (2) fines associated with the dolomitic drainage stone. The relative abundance of dolomite to quartz plus feldspar in the cover soils can be compared to that of the inorganic clog material to estimate the contribution of the drainage-stone fragments to the detrital load of the leachate. This calculation shows that some inorganic clog samples are enriched in dolomite relative to the cover soils, whereas other samples contain no dolomite (Table 3). Some areas of the KVL leachate-collection system may contain a higher quantity of dolomitic fines in the drainage layer because of the initial delivery of the stone. Dolomitic fines typically accumulate at the bottom of the delivery truck and tend to concentrate in the immediate area where the load of stone was dumped prior to spreading by the bulldozer (Fleming et al. 1999).

4. Conclusions

There is no compelling evidence that dolomitic stone is unsuitable for drainage layers in southern Ontario landfills, provided that careful attention is paid to proper design and construction of the leachate-collection systems. Leachate chemistry for landfills in southern Ontario and the United Kingdom and physical observations for exhumed samples of dolomitic drainage stone from the Keele Valley (southern Ontario) landfill and mesocosm simulators suggest that dolomite does not experience significant dissolution in landfill leachate-collection systems. Landfill leachates are typically supersaturated with respect to carbonate minerals but undersaturated with respect to gypsum. These results provide another reason to limit the disposal in landfills of construction and demolition waste containing gypsum (i.e.,

wallboard). Dissolution of gypsum will increase the calcium content of leachates and contribute to calcite-rich clog formation within the leachate-collection system. However, since most modern landfill operations already separate construction debris from MSW before disposal, this scenario is likely to be of diminishing importance in the future.

Sizeable quantities of secondary calcite had formed about the dolomitic drainage stone, both in the Keele Valley Landfill (KVL) and in the mesocosm experiments. The neoformed calcite from the KVL drainage layer had enveloped and cemented a significant quantity of detrital fines (dolomite, quartz, feldspar) present in the pore spaces of the collection system adjacent to the drainage stones. Such fines can contribute to the clogging of the leachate-collection system by providing additional sites for heterogenous nucleation of calcite and by physically occluding pore throats between the drainage stones. Use of a geotextile filter immediately above the leachate-collection system will help to minimize the quantity of inorganic fines washed into the drainage layer from landfill waste and daily cover soils. The initial fines content of the drainage stone must also be controlled through careful unloading and spreading practices during initial placement.

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6. References

- Bennett, P.J. 1998. The stable isotopic characterization of carbon and water cycling in municipal solid waste landfills. M.Sc. dissertation, The University of Western Ontario, London, Ont.
- Brune, M., Ramke, H.G., Collins, H.J., and Hanert, H.H. 1991. Incrustation processes in drainage systems of sanitary landfills. *In* Proceedings of the 3rd International Landfill Symposium, Sardinia, Italy. pp. 999–1035.
- Daniel, D.E., and Koerner, R.M. 1993. Quality assurance and quality control for waste containment facilities. U.S. EPA/600/R-93/182, U.S. Environmental Protection Agency, Washington, D.C.
- Desrocher, S., and Sherwood-Lollar, B. 1997. Isotopic and geochemical characterization of landfill methane. *In* Environmental geology of urban areas. Edited by N. Eyles. Geological Association of Canada, Toronto, Ont., pp. 365–371.
- Fleming, I.R., Rowe, R.K., and Cullimore, D.R. 1999. Field observations of clogging in a landfill leachate collection system. *Canadian Geotechnical Journal*, **36**: 685–707.
- Gartner-Lee Limited. 1990. Beare Road site closure report. Vol. 1: A review of monitoring data at Beare Road Landfill. Gartner-Lee Limited, Markham, Ont.
- Herman, J.S., and White, W.B. 1985. Dissolution kinetics of dolomite: effects of lithology and fluid flow velocity. *Geochimica et Cosmochimica Acta*, **49**: 2017–2026.
- McAulliffe, C. 1971. GC determination of solutes by multiple phase equilibration. *Chemical Technology*, **1**: 46–51.
- MOEE. 1994. Incorporation of the reasonable use concept into MoEE groundwater management activities. Ontario guideline B7 (formerly 15-08) Ministry of Environment and Energy (now Ministry of Environment), Toronto, Ont.
- Niemann, W.L., and Hatheway, A.W. 1997. Effect of variable-pH landfill leachate on a carbonate rock aggregate. *Environmental and Engineering Geoscience*, **3**: 423–430.
- Owen, J.A., and Manning, D.A.C. 1997. Silica in landfill leachates: implications for clay mineral stabilities. *Applied Geochemistry*, **12**: 267–280.
- Plummer, L.N., Prestemon, E.C., and Parkhurst, D.L. 1991. An interactive code (NETPATH) for modeling net geochemical reactions along a flow path. U.S. Geological Survey, Water Resources Investigation Report 91-4078, Reston, Va.
- Rowe, R.K. 1998. From the past to the future of landfill engineering through case histories. *In* Proceedings of the 4th International Conference on Case Histories in Geotechnical Engineering, St. Louis, Mo., pp. 145–166.
- Rowe, R.K., Fleming, I., Cullimore, R., Kosaric, N., and Quigley, R.M. 1995a. A research study of clogging and encrustation in leachate-collection systems in municipal solid waste landfills. Final report to the Interim Waste Authority. Geotechnical Research Centre, University of Western Ontario, London, Ont.
- Rowe, R.K., Quigley, R.M., and Booker, J.R. 1995b. Clayey barrier systems for waste disposal facilities. E&FN Spon, London, U.K.
- Rowe, R.K., Fleming, I.R., Armstrong, M.D., Cooke, A.J., Cullimore, D.R., Rittmann, B.E., Bennett, P.J., and Longstaffe, F.J. 1997. Recent advances in understanding the clogging of leachate collection systems. *In* Sardinia 97, Proceedings of the 6th International Landfill Symposium, Sardinia, Italy. pp. 383–390.
- Salisbury, F.B., and Ross, C.W. 1992. Plant physiology. Wadsworth Publishing Company, Port Elizabeth, South Africa.