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Field studies of the leachability of aged brown coal ash

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Abstract

The environmental management of ash produced from the brown coal power stations of the Latrobe Valley region of Australia has been studied. Current practice consists of slurring fly and bottom ash, a short distance to an ash disposal pond. However, storage facilities are approaching capacity and alternative ash management strategies are required in the near future. Initially, the ash produced within the power stations is known to possess a large soluble mass, which can leach rapidly to generate a saline leachate with minor trace metal content. After slurring and deposition within the ash pond, it has been demonstrated that the soluble mass is significantly lower and the ash can be considered as aged or "leached" ash — a more benign waste that meets the criteria for fill material. In order to assess the long-term behaviour of the leached ash and its suitability for co-disposal in engineered sites within overburden dumps, two field cells were constructed and monitored over a period of 1 year. Each cell was 5 × 5 m in area, 3-m deep and HDPE lined with a coarse drainage layer and leachate collection pipe. The first cell only collected natural rainfall and was known as the Dry Cell. The second cell had an external tank of 5000 l installed (200-mm rainfall equivalent) and water was spray-irrigated regularly to simulate higher rainfall and accelerate the leaching process. The cumulative inflow and outflow for each cell has been calculated using a linear relationship and the leachate quality was monitored over time. The results demonstrate that the ash behaves as an unsaturated porous material, with the effect of evaporation through the profile being dominant and controlling the production of leachate. The leachate quality

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was initially moderately saline in both cells, with the concentration dropping by nearly 95% in the Wet Cell by the end of the field study. The leachate chemistry has been analysed using the PHREEQC geochemical model. The log activity plots of various species suggest the mineralogical controls on these species in leachate. The full results from this study are presented. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Leachability; Aged ash; Field studies; Brown coal ash

1. Introduction

The Latrobe Valley region of Victoria, Australia, mines in excess of 50×10^9 kg of low rank brown coal per year from three large open cut operations for use in the production of electricity in four main generating complexes. These include the Hazelwood (1600 MW e), Yallourn (1450 MW e), Loy Yang Power (2000 MW e) and Edison Mission Energy facilities (1000 MW e, part of the Loy Yang complex). This leads to the creation of over 550×10^6 kg of solid wastes per year, consisting mostly of precipitator (fly) ash, partially burnt coal particles (char), furnace (bottom) ash, salts, sand and clay minerals [1]. Presently, the ash produced in the power stations is mixed with water and transported to an ash disposal pond in a high water-to-ash ratio slurry. However, the capacity of existing ash disposal ponds are limited and the feasibility of expanding the storage capacity or establishing new facilities is restricted.

The historical research on ash leaching behaviour was performed by the former State Electricity Commission of Victoria (SECV) during the late 1960s through mid 1980s. This work generally concentrated on engineering aspects and scaling problems in the hydraulic ash transport system, however, and not those relevant to ash disposal behaviour [2]. All chemical analyses of ash were performed on a total concentration basis, and meeting Environment Protection Authority Victoria (EPAV) criteria for waste management of the ash often presented substantial environmental, engineering and economic considerations [3–5].

At the Loy Yang Power complex, there has been active and ongoing research over recent years to identify alternative ash disposal, management and possible utilisation strategies. The strategy adopted to date has been to excavate ash material from the ash pond and develop specific disposal sites within the Loy Yang Overburden Dump. However, to determine the appropriate level of environmental and engineering design requirements, research has been directed at determining the behaviour of ash excavated from the ash pond in ways that more accurately simulate the field conditions expected at disposal sites.

Current EPAV methodology stipulates that all analyses of contaminated soils and wastes only need consider the acid-extractable fractions and not those included within the silicate matrix or otherwise of limited availability to the environment [6].

Thus, current research has been aimed at determining the degree to which the ash has been leached through the hydraulic ash transport system, the EPAV criteria for assessing the requirements for the ash as a waste material, and ash behaviour under in situ field conditions at the Loy Yang site. The latter two aspects of this research are presented.

2. Ash mineralogy and characterisation

The mineralogy of the ash produced in the Latrobe Valley has been studied by several workers, with the most notable contributions being that of Black et al. [1], Black [2,7], Bone and Schaap [8,9]; Schaap [10] and Deed [11–13]. The engineering and soil properties of Latrobe Valley ash were studied extensively and reported by Peterson et al. [14]. There are known to be physical and chemical differences between the ash produced at Loy Yang, Hazelwood and Yallourn, thought to be due to the variation in coal quality between the sites and operational differences [1]. A typical ash analysis is given in Table 1.

The main ash forming minerals that have been identified to date in Latrobe Valley ashes include thenardite (Na_2SO_4), halite (NaCl), periclase (MgO), haematite (Fe_2O_3), magnetite (Fe_3O_4), lime (CaO), anhydrite (CaSO_4), alpha-quartz ($\alpha\text{-SiO}_2$), alumina (Al_2O_3) and minor variations of calcium and magnesium species [1,2,7,10]. At the Hazelwood complex, other minerals identified in ash pond sediment include gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) and calcite (CaCO_3 , formed from exposure to carbon dioxide in the atmosphere) [2]. It should be noted that there is a degree of variability in mineral compositions in Latrobe Valley ash deposits.

The ash from Latrobe Valley power stations generally has poor pozzolanic characteristics due to higher soluble salt loads [1,14]. There has been some detailed studies of trace metals in Latrobe Valley ashes (e.g. Refs. [8–10]); however, the strong alkalinity of Latrobe Valley ash effluents limits the solubility of most metals in the leachate [2].

The heterogeneity of the brown coal excavated from the different open cuts, as well as variability within a particular open cut, leads to a correspondingly wide range of these major constituents in the ash. However, the proportions of these different constituents tend to control the chemistry of ash leachate observed in slurry systems and the ash ponds at each site [15]. All power stations maintain a moderate to strongly alkaline ash disposal system.

Table 1
Comparison of Latrobe Valley brown coal ashes (%) [1]
LOI—Loss on ignition.

Species	Loy Yang	Yallourn	Hazelwood
SiO_2	60.4 ^a	1.4	6.6
Al_2O_3	13.3 ^a	2.1	1.8
Fe_2O_3	8.5	24.5	8.7
TiO_2	1.7	0.1	0.2
K_2O	1.2	0.4	0.4
MgO	2.2	18.0	18.8
Na_2O	2.1	11.0	4.5
CaO	1.0	12.3	28.4
SO_3	3.4	21.7	15.6
Cl	< 0.1	< 0.1	3.4
LOI	7.6	8.2	11.7

^aHighly variable.

Research in the Latrobe Valley and internationally has demonstrated that ash leaches rapidly at first, and then approaches a steady state leachate quality (e.g. Refs. [2,7,16–19]). This behaviour is similar to landfills and to many other waste forms. Farquhar [16] presented the change in leachate quality and the leachable mass within a waste form conceptually as shown in Fig. 1. However, it is the point at which the ash is rendered effectively leached and aged that is critical in long term management of ash disposal sites. At this point, the potential for adverse environmental impacts becomes minimal.

There are several processes that act simultaneously to determine leachate production, chemistry and behaviour. These include the extent of advective flow through an ash, possible diffusion from the ash matrix to surrounding leachate, and geochemical conditions of the leachate and ash. Transport of an ash slurry within a pipeline to a disposal pond would create strong advective leaching, whereas within the ash pond the leaching would be more influenced by diffusion, mineral solubility and absorption of atmospheric carbon dioxide. Thus in Fig. 1, Point A would represent the rapid advective leaching of highly soluble surface salts with a subsequent decrease in leachate concentration; while Point B, where the concentration is significantly lower, shows the limited leachate concentration controlled more by diffusive leaching fluxes from within the ash matrix.

The physical properties of the ash exhibit some similarities between the power stations with the characteristic variability expected for the ash. The ash produced is of low density and high porosity, and predominantly of a fine sandy-silt type particles, with approximately 66% less than $75\ \mu\text{m}$ in size [14,20,21]). The in situ dry bulk densities of Latrobe Valley ash are approximately 440 to $600\ \text{kg}/\text{m}^3$, with corresponding saturated densities of 1100 to $1650\ \text{kg}/\text{m}^3$ [14]. Reported values of porosity range from 39.1% to

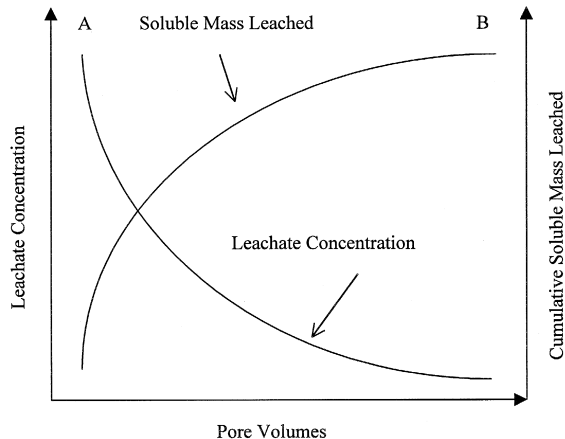


Fig. 1. Change in ash leachate and soluble mass vs. time (adapted from Ref. [16]). Point ‘‘A’’ is the maximum concentration of a leachate species, and is controlled by mineral solubility, geochemical conditions, ash morphology and other factors. Point ‘‘B’’ represents the effective solubility of a particular species within the ash, and is controlled by ash morphology, coal quality, combustion and boiler characteristics.

80.9% [1,21]. The specific gravity of ash has been found to range from 1.2 to 3.25, with the average approximately 2.34 [14,21,22].

3. Leached ash concept

It has been established by previous workers that the hydraulic transport system and deposition within a saturated ash disposal pond significantly reduces the level of soluble salts within the ash [7–9,21,23]. The total solubility of dry precipitator ash produced within Latrobe Valley power stations has been reported to range from 16% to as high as 39% [2,7], while the total solubility for ash derived from ash pond sediments has much lower solubility ranging from 4% to 11% [2,7].

To demonstrate the relatively low concentrations in ash derived from disposal ponds, a detailed study was undertaken at Loy Yang to assess the degree of ash solubility after transport and deposition and the level of trace metals in the ash according to EPAV methodology. The hypothesis was to determine if the ash could be considered as acceptable fill material or *leached ash*. This study was reported in Mudd et al. [5] and Mudd [21] and is briefly summarised below.

In April 1995, a series of 12 samples of ash were taken from the Loy Yang Ash Pond delta and tested for trace metal levels using EPAV techniques. The ash samples were analysed for barium, cadmium, chromium, cobalt, molybdenum, nickel, tin, and mercury. Results are summarised in Table 2, including previous analyses by the SECV (which were on a total solid concentration basis). A further series of 12 samples from approximately the same sites on the Loy Yang Ash Pond delta was obtained and analysed in November 1995, with the results presented in Table 3.

The three samples from the November 1995 series with the highest barium concentration (which includes the sample with the highest mercury concentration) were selected for more aggressive leaching tests. A dual sequential batch leaching procedure was adopted using both distilled water and acidified eluents, based on the standards ASTM D 4793-93 and D 5284-93 [24]. These samples were also analysed for their major constituents and trace metals based on a total concentration technique. The results from the batch tests gave an average solubility of 1.77% and 2.24% of total mass for distilled water and the acidified solution, respectively [21]. The only trace metal leached from each batch test was barium at about 0.8% (of total barium in the ash) [21].

Table 2

Leached ash study April 1995 (mg/kg) [21]

Only one ash sample exceeded the EPAV criteria for Ba and Hg.

	Ba	Cd	Cr	Co	Mo	Ni	Sn	Hg
Minimum	18	< 0.5	7	< 1	< 4	1	< 4	0.2
Maximum	420	< 0.5	110	18	< 4	20	< 4	2.1
Average	140	< 0.5	30	7.3	< 4	11.5	< 4	0.81
SECV	–	0.4	150	80	25	94	44	0.21
EPAV	400	5	250	50	40	100	50	2

Table 3

Leached ash study November 1995 (mg/kg) [21]

Three ash samples exceeded the EPAV criteria for Ba and one equalled the criteria for Hg.

	Ba	Cd	Cr	Co	Mo	Ni	Sn	Hg
Minimum	78	0.1	18	6	< 5	11	10	0.16
Maximum	550	1.3	57	19	12	32	39	2.0
Average	264	0.77	33	11	8.3	20	24	0.98
EPAV	400	5	250	50	40	100	50	2

At the Loy Yang complex, it was assessed that the ash, after being hydraulically slurried, deposited and leached within the Ash Pond, becomes a fill material according to EPAV criteria, and has much lower potential to lead to adverse environmental consequences [21,22]. Thus, the ash excavated from the Loy Yang Ash Pond is accordingly described as *leached ash*, to clearly distinguish it from *unleached* precipitator ash (which remains a prescribed waste by EPAV).

However, the batch testing methodologies are designed to indicate the total solubility and availability, and not the actual time-dependent processes in disposal scenarios [25,26]. The use of total concentration data cannot provide any indication as to the rate and extent of mobilisation of any of the elements when the wastes are subject to chemical weathering [25,27]. The different factors that may influence release in disposal sites such as engineered landfills, or in utilisation scenarios such as road sub-base materials or cement mixes, include advective transport, diffusive transport, and geochemical conditions (especially important for trace metal behaviour) [26].

4. Field leaching trials

4.1. Engineering design and monitoring

At the Loy Yang complex and elsewhere in the Latrobe Valley, leached ash is currently excavated from the deltas of disposal ponds and is placed in suitably located and engineered sites approved by EPAV. To verify the long-term behaviour of leached ash under unsaturated conditions, trial-leaching cells were constructed at Loy Yang in late June 1997. Due to the climate of the region, two individual cells were constructed with one cell remaining open to rainfall while the second cell had an external 5000-l water tank installed for regular irrigation, equivalent to 200 mm of rainfall. The cells were constructed in a special purpose ash platform located above the maximum water level of the pond and within the southwestern confines of the Ash Pond catchment. The two cells were, respectively, named the Dry Cell and the Wet Cell. By building two separate cells, natural leaching rates and flow behaviour could be established while an accelerated leaching test could be conducted on the second cell by adding water regularly to reach much higher pore volumes of leaching.

The design and construction of each cell was identical, with an area of 5 m by 5 m at the surface and a height of 3 m. The liner used was 2-mm thick High Density

Polyethylene plastic (HDPE) and a 75-mm drainage layer was installed at the base consisting of sub-rounded aggregate at 3 to 5 mm diameter. A layer of geotextile (Bitton Cloth) was used to separate the ash layer from the drainage layer and prevent possible clogging and fine particle migration within each cell. The potential for puncture of the HDPE liner is low, using a method by Ref. [28], given the very high factor of safety calculated for the design of the cells.

The drainage layer, and possibly the geotextile, can act as a capillary break. The amount (and rate) of leachate emanating from the base of an ash layer will depend on the permeability and moisture condition of underlying soils or materials. In the field situation, disposal sites are lined with unsaturated clays at the time of placement, compared to the cells, which include the geotextile, drainage layer and HDPE. Thus, the cells provide a worse case scenario for leachate generation. This is due to the low permeability of clays even after they are saturated by percolating leachate (leading to localised ponding in the ash layer); whereas the drainage layer has a very high permeability and no ponding can occur due to rapid lateral discharge.

The density of ash within each cell was generally similar to that found in current and proposed ash dumpsites within the Loy Yang Overburden Dump [21]. A perforated leachate collection pipe was installed within the coarse aggregate layer and extended beyond the liner to a collection tank. The tank volume of 300 l was sufficient to hold a maximum rainfall event typical of the Latrobe Valley. The Wet Cell had an array of three lines of irrigation spray jets installed, sufficient to achieve an irrigation rate of about 11 mm/h, making the total time for one tank of water to irrigate the Wet Cell approximately 18 to 20 h. The water used to refill the Wet Cell tank was derived from the Loy Yang High Quality Water system, and was tested regularly for water quality. The tank was allowed to irrigate the Wet Cell approximately once every 2 weeks, and both the leachate flow rate emanating from the collection pipe and the total volume collected per event was monitored very closely.

The operational and monitoring period of both cells lasted from July 1997 to August 1998. A detailed calculation of total inflow and outflow was established for each cell, whereby rainfall, external water and total outflow of leachate was monitored. Samples of leachate from each cell were obtained approximately every 2 weeks (or more frequently when required), and analysed for a complete suite of cations, anions, trace metals, pH, salinity, and redox conditions of the leachate. A series of ash samples was taken at the time of construction of each cell, and further surface samples and series of ash samples at various depths were taken approximately every 3 to 4 months. In combination with the inflow, outflow, ash and leachate analyses, a detailed mass balance was calculated for each cell.

4.2. Climate data and influent water quality

The monitoring of meteorological data was performed by the Loy Yang weather station, situated close to the northwestern corner of the Ash Pond. The parameters monitored were rainfall, daily pan evaporation (Class A pan, monitored by Loy Yang Power staff), minimum and maximum temperature, minimum and maximum relative humidity, and wind speed. Climate data is summarised in Table 4.

Table 4

Average monthly climate data for Loy Yang July 1997–August 1998

1997–1998	July	Aug.	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.
Rainfall	43.2	57.2	55.2	54.4	69.6	20.7	50.4	62.2	7.2	29.9	32.4	51.2	40.8	27.2
Pan evaporation	16.9	64.2	58.9	121	123	237	102	149	165	57.7	53.2	32.7	27.0	32.0
Maximum temperature	n.d.	n.d.	n.d.	20.0	22.0	23.9	27.2	26.7	24.2	n.d.	15.6	13.0	11.3	14.0
Minimum temperature	n.d.	n.d.	n.d.	8.6	10.5	11.9	15.0	12.9	11.8	n.d.	8.8	5.9	4.1	6.6
Maximum RH	n.d.	n.d.	n.d.	95	94	93	94	94	94	n.d.	96	99	99	99
Minimum RH	n.d.	n.d.	n.d.	54	54	47	48	45	49	n.d.	74	72	77	69
Wind speed	n.d.	n.d.	8.2	10.8	9.9	11.5	10.8	9.7	9.6	9.7	9.4	7.3	8.1	7.3

Rainfall and pan evaporation (mm); maximum/minimum temperature (°C); maximum/minimum relative humidity (RH) (%); wind speed (km/h).

n.d. is no data (due to equipment malfunction). Available data is taken from daily pan evaporation.

Samples of rainfall water quality were taken, as well as several samples of the external tank on the Wet Cell. All samples had a salinity approximately less than 50 mg/l, with only minor amounts of aluminium and zinc [21].

4.3. Dry Cell results

4.3.1. Physical properties

The porosity and density characteristics of the Dry Cell were determined in July 1998 and are presented in Table 5. The values are similar to those measured for the nearby Ash Dumpsite, which the Dry Cell was designed to simulate. There appears to be increasing moisture content and degree of saturation with depth. This phenomenon has been observed within the Ash Dumpsite [21] and is thought to be influenced by self-weight consolidation, drainage of excess initial pore water, equilibration of pore pressures and the capillary break effect of the drainage layer [22].

4.3.2. Cumulative flows of the dry cell

The cumulative flows of the Dry Cell consist of rainfall, evaporation, leachate generation and change in moisture storage within the ash profile. Over the full year of

Table 5

Geotechnical index properties of the Dry Cell

SC: South central corner; NC: North central corner.

Site and depth	Moisture content (%)	Density (kg/m ³)		Void ratio	Porosity (%)	Degree of saturation (%)
		Wet	Dry			
SC Top	85.9	1111	598	2.92	74.5	68.9
NC Top	86.2	847	455	4.15	80.6	48.7
NC 1.4 m	86.4	1027	551	3.25	76.4	62.3
NC 2.6 m	96.2	1153	588	2.98	74.9	75.5

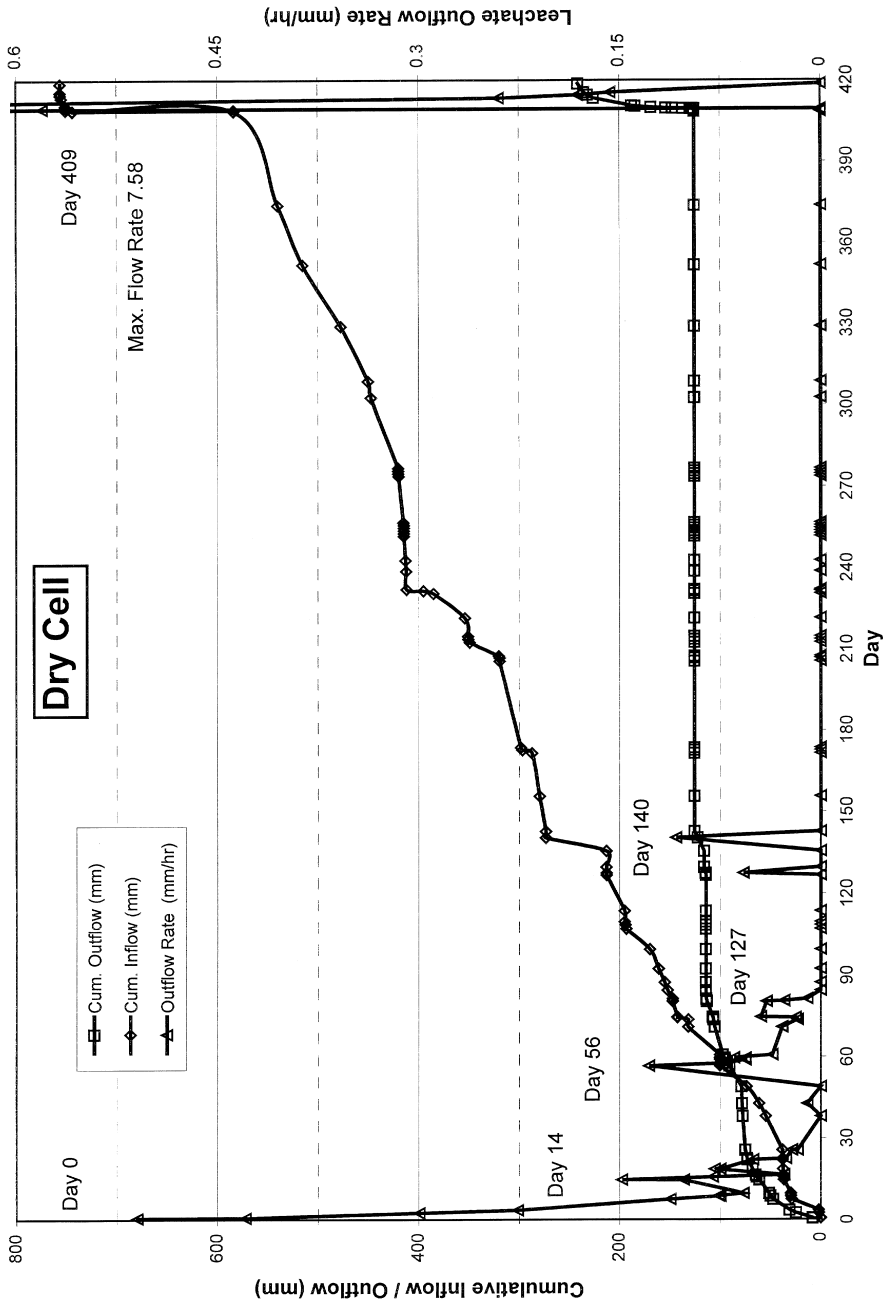


Fig. 2. Hydraulic performance of the Dry Cell.

the study, only minor quantities of leachate were generated, almost all of which was within the first 3 months of operation. The flow rates of leachate were also quite low. The results are given in Fig. 2.

Due to the unusually low quantity of rainfall over the period from July 1997 to July 1998, there was very little or no discharge of leachate from the Dry Cell since mid-September (1997). The only occurrence of leachate flow after this time was during a period of high rainfall in early November (1997), which itself produced only a very small quantity of leachate (less than 12 mm compared to approximately 60 mm of rainfall). The remaining rainfall that has fallen on the Dry Cell has been held in soil moisture storage, part of which was later lost to evaporation.

For the first few weeks of operation, the estimated outflow from the Dry Cell slightly exceeds the inflow from rainfall (on the basis of measured data). This is likely due to the sparse nature of the flow rate data measured during this time. The total outflow is estimated as the area under the flow rate vs. time graph by linear interpolation. The tendency is to overestimate the outflow since a linear flow rate is assumed between measured data points. The flow may also be influenced by self-weight consolidation of the ash within the cell and equilibration of pore pressures leading to minor leachate generation. Such behaviour has been observed at the current Ash Dumpsite at Loy Yang [22].

To obtain further samples of leachate from the Dry Cell, the irrigation tank was shifted from the Wet Cell and allowed to irrigate the Dry Cell in mid-August 1998. A total of 196 mm was irrigated over a period of 10 h, and three leachate samples were obtained for analysis over succeeding days. The estimated volume of leachate collected was approximately 115.6 mm, just over half of the input volume.

Overall, there is a net evaporation of water in the Dry Cell. By August 1998, a total of approximately 755.8 mm of rainfall (including the extra tank water) entered the cell, whereas only 241.2 mm of leachate has been calculated to discharge from the cell. Given that the pore volume of the Dry Cell is approximately 2298 mm, the Dry Cell has not reached any significant stage of field leaching, with about one tenth of a pore volume of leachate generated to date.

4.3.3. Leachate chemistry of the Dry Cell

Water quality of leachate discharged from the Dry Cell was measured in the early months of its operation and during irrigation with the final tank of water in August 1998. A total of 15 samples was collected and analysed for major ions and a suite of trace elements (Al, As, B, Ba, Cu, Co, Cr, Cd, Hg, Mo, Ni, Pb, Se, Sn, Zn). There were no

Table 6

Range in leachate quality of the Dry Cell throughout the trial: major ions (mg/l)

	pH	TDS	SO ₄	Cl	HCO ₃	CO ₃	Na	K	Ca	Mg	SiO ₂
Minimum	7.6	8700	2600	630	46	16	2100	70	93	22	7.4
Maximum	9.3	17,000	13,000	1100	210	64	5800	130	210	220	11
Average	8.5	10,600	6222	773	67	44	3556	98	119	42	9.3

Table 7

Range in leachate quality of the Dry Cell throughout the trial: trace elements ($\mu\text{g}/\text{l}$)

	Al	As	B	Ba	Mo	Se	Zn
Minimum	60	3	900	30	< 0.01	95	< 10
Maximum	1400	12	3400	180	390	3800	80
Average	304	8	2386	120	288	697	12

samples between November 1997 and August 1998 due to the absence of leachate from the cell. The results are summarised in Tables 6 and 7.

The initial samples of leachate gave only slightly oxidising conditions (around 25 to 45 mV). However, this was quickly increased to much stronger oxidising conditions with the redox state stabilising for the remainder of the study around 260 to 280 mV [21].

The leachate is moderately to highly saline, being dominated by sodium, sulphate and chloride. The leachate is moderately alkaline, with the pH varying around 8.5 to 9. The level of iron in Dry Cell leachate was generally less than 0.2 mg/l. Most trace elements were either very low or below detection limits, which would be due to the alkaline nature of the leachate. However, the notable exceptions are boron, barium, molybdenum and selenium, which consistently register in the leachate.

The following metals were consistently near or below detection limits ($\mu\text{g}/\text{l}$): Cd < 1, Co < 10, Cr < 10, Cu < 10, Pb < 10, Hg < 1, Ni < 10 and Sn < 10.

There has been no discernible pattern in the data apart from being of similar magnitude. The small leachate sample from November 1997 gave a much higher salinity. This may be due to the long time duration between samples (i.e. a higher residence time), which allowed more chemical constituents to dissolve or diffuse from the ash matrix. This extra time allowed a progressively lower moisture content within the ash profile due to evaporation, effectively concentrating the leachate (suggested by the higher chloride level of this sample). With the advent of a high rainfall week, this water was flushed out from the ash through advective flow with minimal mixing due to the low volumes. There does not appear to be any increase in trace element levels with this sample, only an increase in salinity due to higher sulphate, sodium and chloride.

4.3.4. Ash quality of the Dry Cell

During the full year study, ash samples were collected from the Dry Cell and analysed for major and trace elements. These included surface samples taken approxi-

Table 8

Range of ash quality of the Dry Cell throughout the trial: major elements (% DB)

AI: Acid insoluble; LOI: Loss on ignition.

	SO ₄	Cl (mg/kg)	Na	K	Ca	Mg	Fe	AI	AI	LOI
Minimum	0.18	570	0.58	0.041	2.2	3.4	1.9	1.6	49	13
Maximum	1.20	1800	1.90	0.160	4.0	7.1	7.4	4.8	70	30
Average	0.66	963	1.29	0.113	3.0	4.7	3.9	2.9	62	20.7

Table 9

Range of ash quality of the Dry Cell throughout the trial: trace elements (mg/kg)

	As	B	Ba	Cd	Cr	Co	Cu	Hg	Mo	Ni	Pb	Se	Sn	Zn
Minimum	2.6	110	160	< 0.1	27	16	27	0.56	< 5	33	< 5	< 5	< 1	64
Maximum	10	250	650	0.4	62	420 ^a	64	2.50	23	130	15	12	< 5	150
Average ^b	7.4	169	304	0.23	36	22	40	1.29	9.6	40	11	8.2	< 5	84

^aStatistical outlier, all other values < 30 mg/kg.^bExcluding values below detection.

mately every 3 months, and two boreholes of vertical samples taken every 6 months. A total of 26 samples were collected. The results are summarised in Tables 8 and 9 (dry basis).

There is no discernible pattern in the ash chemistry, except for the typical variability found in ash samples. The average levels of trace metals are again within EPAV criteria for consideration as fill material, with only select samples near or above the criteria for certain metals (such as barium with 11 samples and mercury with five samples). This further demonstrates the lower leachability of ash excavated from the Loy Yang Ash Pond.

4.4. Wet Cell results

4.4.1. Physical properties

The porosity and density characteristics of the Wet Cell were determined in July 1998 and are presented in Table 10. The values are similar to those measured for the nearby Ash Dump site and the Dry Cell. Similar to the Dry Cell, the moisture content and degree of saturation appear to increase with depth.

4.4.2. Cumulative flows of the Wet Cell

The cumulative flows for the Wet Cell also demonstrated typical unsaturated soil behaviour, although higher inflow rates took place into this cell. After construction in late June 1997, the Wet Cell exhibited similar flow rates to the Dry Cell from the leachate discharge pipe. The leachate flow rate decreased over the first few weeks as the ash consolidated. However, as the external water supply was added, the peak flow rate

Table 10

Geotechnical index properties of the Wet Cell

C: Central corner; NW: North western corner.

Site and depth	Moisture content (%)	Density (kg/m ³)		Void ratio	Porosity (%)	Degree of saturation (%)
		Wet	Dry			
C Top	86.0	832	447	4.23	80.9	47.6
NW Top	81.3	812	448	4.22	80.9	45.0
NW 1.4 m	85.1	1080	583	3.01	75.1	66.1
NW 2.6 m	93.7	1127	582	3.02	75.1	72.6

of leachate increased with each addition of water. The decrease in the leachate flow rate following each event was rapid. The peak flow rate for each of the first few events of adding water showed an increasing trend. The results are presented in Fig. 3.

In general, the variation in peak flow rate for each event of adding water can be explained on the basis of unsaturated flow mechanics. For an unsaturated soil system, the hydraulic conductivity of a soil depends on its moisture content, and thus the velocity of the moisture movement. As the moisture content of the ash profile increases with the addition of water, the hydraulic conductivity increases and higher flow rates through the ash profile can be achieved. This is particularly highlighted by the lower flow rates achieved through the summer months (days 150 to 220, i.e. 29 November 1997 to 7 February 1998).

The event on day 204 (22 January 1998) demonstrates that despite a full tank of 200 mm added to the cell, the overall flow rate was very low, indicating that the majority of the water was held in soil storage due to the much drier profile. The next two events (day 211, 29 January 1998; and day 229, 16 February 1998), however, achieved two of the highest outflow rates recorded to that point in time. The soil moisture deficit of the ash profile had been reduced by the influx of water on day 204, allowing water to flow through the ash profile relatively rapidly.

For the first few months of operation, there were some occasions where the estimated outflow from the Wet Cell slightly exceeded the inflow from rainfall and the source tank. This may be influenced by the use of linear interpolation in the calculating the volume of leachate outflow, which is the area under the outflow rate vs. time (cf. Fig. 3). Similar to the Dry Cell, the flow may also be influenced by self-weight consolidation of the ash within the cell and equilibration of pore pressures leading to minor leachate generation. This is thought to explain the early data for the Wet Cell, where the outflow marginally exceeds inflow at some points.

For the last 2 months of actively operating the Wet Cell, an accelerated program of irrigation was undertaken, where up to four tanks per week were added. Again, typical unsaturated soil behaviour is evident. As the second or third tanks of water were added to the cell, the peak outflow rate also increased, indicating a higher permeability due to a higher moisture content profile within the ash. For the final weeks in July 1998, the outflow rate almost approached the inflow rate, suggesting that the ash was behaving as a saturated soil where the hydraulic conductivity may have been controlled by saturated behaviour.

Overall, there is a net evaporation of water in the Wet Cell. A total of approximately 7540.6 mm of combined rainfall and source tank water has entered the cell, whereas only 5555.2 mm of leachate has been calculated to discharge from the cell. Given the pore volume of the Wet Cell at approximately 2340 mm, this represents about 2.4 pore volumes of active leaching.

Some important patterns to note are the respective gradients of the Inflow and Outflow curves. During both the Winters of 1997 (about day 120; 30 October 1997) and 1998 (about day 330; 28 May 1998), the ratio of the gradient for each curve is about unity (meaning the quantity of water entering the cell is approximately the same as that discharging), compared to Spring 1997 (about day 200; 18 January 1998) where the ratio begins to decrease. The ratio is lowest during Summer (about day 260; 19 March 1998),

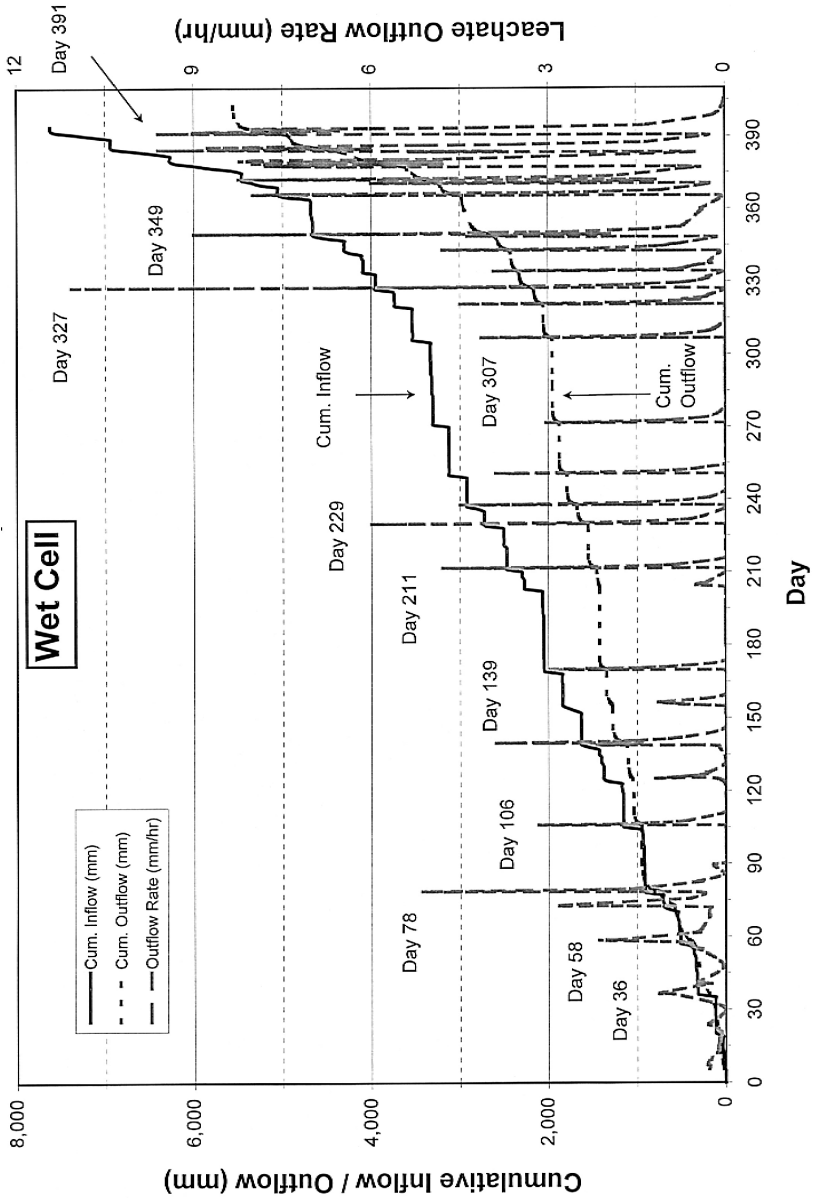


Fig. 3. Hydraulic performance of the Wet Cell.

Table 11
Average leachate quality of the Wet Cell : major ions (mg/l)

Date	pH	TDS	SO ₄	Cl	HCO ₃	CO ₃	Na	K	Ca	Mg	SiO ₂
July–Sept. 1997	9.2	11,000	6780	671	38	68	3467	119	130	26	9.3
Dec. 1997	9.3	7300	4100	300	40	60	2300	85	90	15	10
March 1998	8.3	5700	3800	150	110	2	1800	84	77	9.8	8.7
15 June 1998	9.2	3100	2000	100	30	44	950	46	38	10	7.5
29 July 1998	10.0	1,000	480	28	–	68	330	20	15	2.4	6.5
Decrease (%)	–	91	93	96	–	–	91	83	89	91	–

and begins to increase again during Autumn 1998 (about day 330; 28 May 1998). This is again expected as the evaporative demand during Summer is higher and therefore there is less water available for leachate discharge.

4.4.3. Leachate chemistry of the Wet Cell

A total of 52 leachate samples were collected from the Wet Cell, and results are summarised in Tables 11 and 12 and Figs. 4 and 5. The leachate quality is presented as normalised concentrations with respect to the average leachate concentration between July and September 1997, when concentrations were approximately constant.

The initial leachate obtained was of similar salinity and chemistry to leachate from the Dry Cell, being dominated by sodium, sulphate and chloride and moderately alkaline pH. The same trace metals were also leached at similar levels to the Dry Cell. The same level of salinity was maintained for about 3 months, from whence it continued to decrease until the end of the trial.

The initial samples of leachate had slightly oxidising conditions (around 45 mV). However, this was quickly increased to much stronger oxidising conditions with the redox state stabilising around 300 to 380 mV [21]. As the irrigation rate was increased in July and August 1998, the redox state began to decrease to about 160 to 180 mV [21].

In summary, there was an average decrease of about 90.3% in most major elements between July 1997 and August 1998, especially sodium, sulphate and chloride (which account for the majority of leachate salinity). The level of iron in Wet Cell leachate was always less than 0.5 mg/l. There were some select trace metals leached, namely aluminium, arsenic, boron, barium, molybdenum and selenium. The change in leachate

Table 12
Average leachate quality of the Wet Cell: trace elements ($\mu\text{g/l}$)

Date	Al	As	B	Ba	Mo	Se
July–Sept. 1997	810	9	3300	70	260	810
Dec. 1997	210	6	4900	30	130	2000
March 1998	640	3	7900	< 10	110	1500
15 June 1998	50	3	4300	20	60	218
29 July 1998	460	< 5	4800	< 10	< 10	80
Change (\pm %)	–43.8	–67	+145	–86	–96	–90

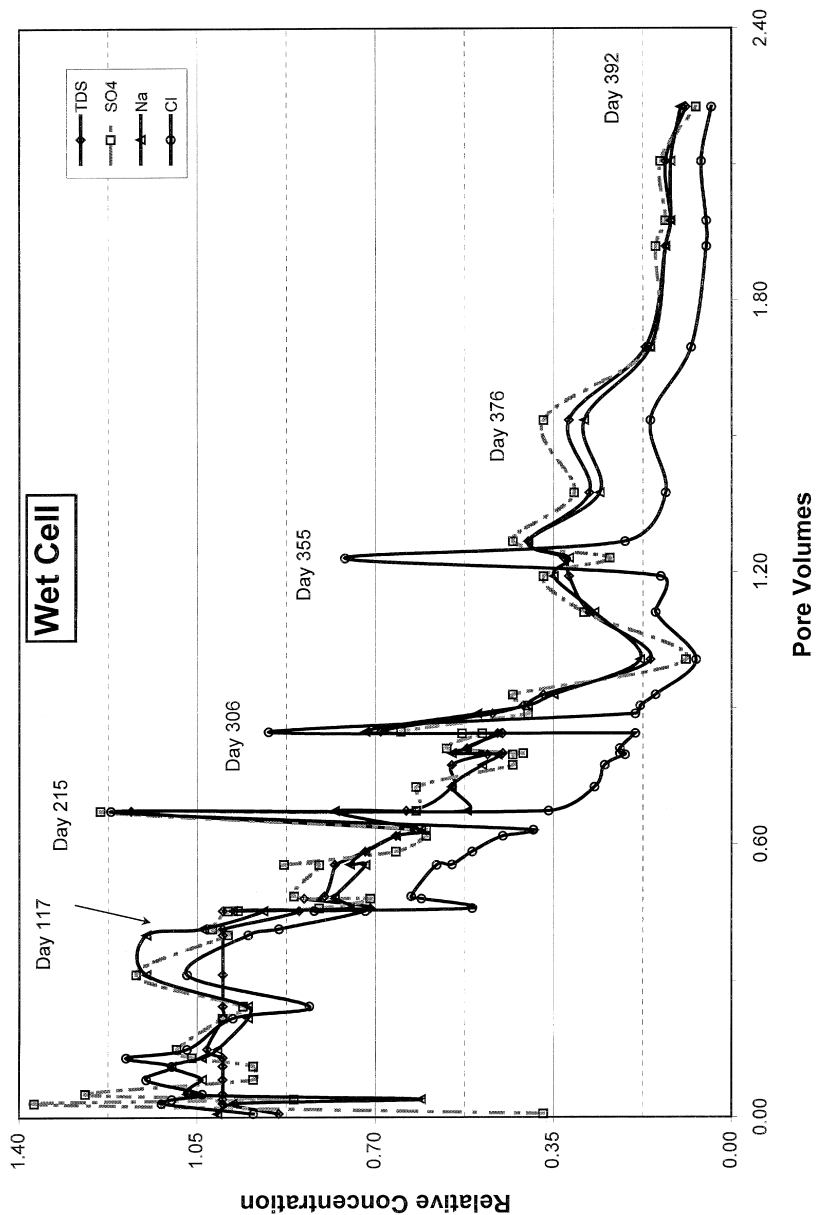


Fig. 4. Leachate quality vs. pore volume: major elements.

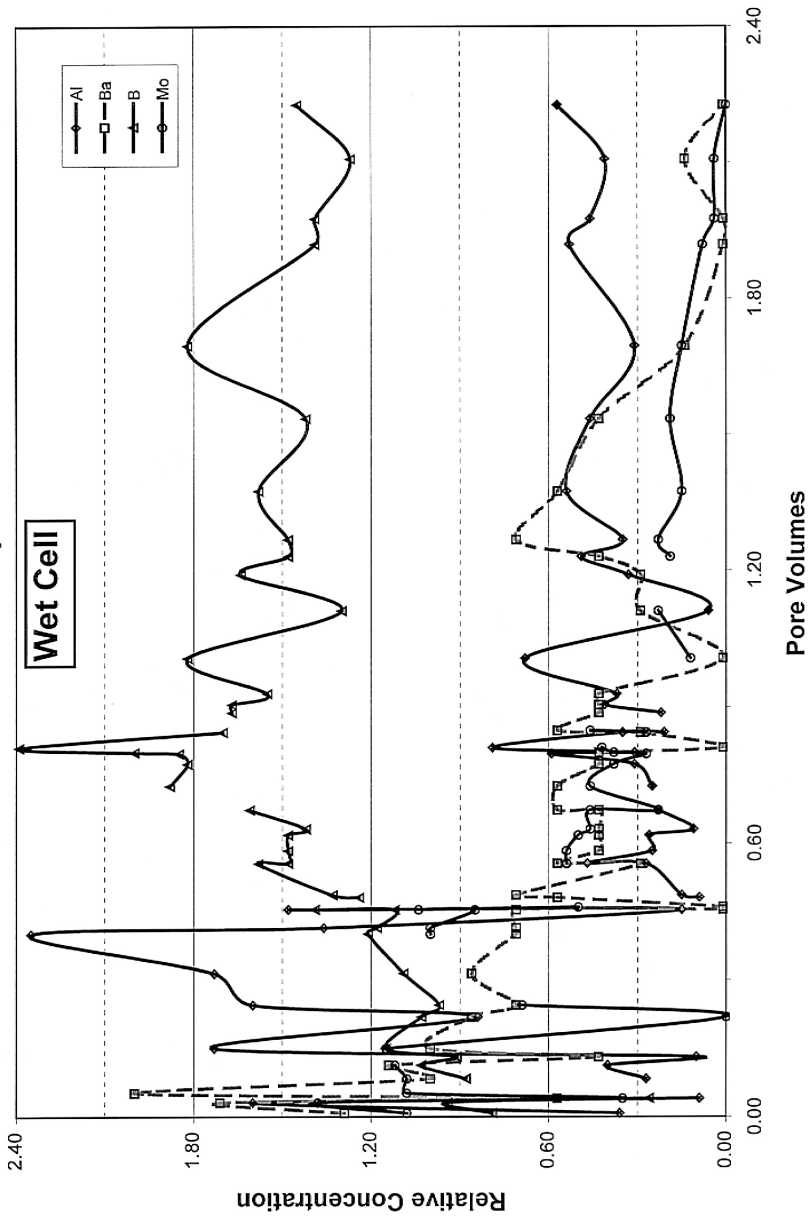


Fig. 5. Leachate quality vs. pore volume: trace elements.

Table 13
Average ash quality of the Wet Cell : major elements (%)

Date	SO ₄	Cl (mg/kg)	Na	K	Ca	Mg	Fe	Al	Al	LOI
July 1997	0.86	728	1.38	0.128	3.8	5.6	3.9	2.9	57	22
Oct. 1997	0.28	110	0.66	0.077	3.0	4.8	4.4	2.9	67	21
Feb. 1998	0.41	129	0.74	0.083	2.8	4.5	3.5	2.6	67	20
May 1998	0.38	905 ^a	1.07	0.116	3.1	4.6	4.7	3.7	61	10
Aug. 1998	0.21	–	0.59	0.070	2.9	4.8	5.4	3.7	62	19
Decrease (%)	76	98 ^b	57	45	–	–	–	–	–	–

^aAverage based on 1800 and < 10 mg/kg.

^bAssuming final Cl of 10 mg/kg.

concentrations over time was not as marked for trace metals as for major elements, except for boron, the concentration of which actually increased over the trial period.

The following metals were consistently near or below detection limits (all µg/l): Cd < 1, Cr < 10, Co < 10, Cu < 10, Pb < 10, Hg < 1, Ni < 10, Sn < 10 and Zn < 10. There were a small number of samples that registered Cr, Co, Cu, Ni and Zn; however these were close to detection limits and are not considered significant.

4.4.4. Ash quality of the Wet Cell

The chemical quality of the leached ash has been determined regularly throughout the operation of the Wet Cell. Due to the higher quantity of water passing through the Wet Cell, active leaching has occurred and a distinct change in ash chemical quality is apparent. The average results of 40 ash samples from the surface and at depth over time are presented in Tables 13 and 14.

4.4.5. Changes in soluble mass

To ascertain the extent of leaching for particular species, the total soluble mass was calculated before and after the 1-year of active leaching, on the basis of dry density of the ash and the concentration of various soluble salts. For the major species sulphate, chloride, sodium and potassium, between 45% and 98% of their soluble mass was leached. For the less soluble species, calcium, magnesium, iron and aluminium, there appears to be no significant change in soluble mass. However, by calculating the mass leached based on concentration of a species in the leachate and the total volume of

Table 14
Average ash quality of the Wet Cell: trace elements (mg/kg)

Date	As	B	Ba	Cd	Cr	Co	Cu	Hg	Mo	Ni	Pb	Se	Sn	Zn
July 1997	9.6	153	180	0.3	33	24	37	1.58	< 5	43	12	< 5	< 5	74
Oct. 1997	4.4	116	570	0.14	44	22	41	1.85	2.2	89	10	< 5	< 5	77
Feb. 1998	11	171	400	< 0.2	34	24	41	0.83	< 5	40	12	< 5	< 5	85
May 1998	8.5	121	455	0.12	36	29	37	1.20	3.9	39	9.2	5.3	3.3	75
Aug. 1998	< 5	128	607	< 0.2	37	25	37	1.45	10 ^a	40	11	< 5	< 5	93

^aExcluding values below detection.

Table 15

Total soluble mass in leached ash and leachate: major elements (kg)

	SO ₄	Cl	Na	K	CO ₃	Ca	Mg	Al	Fe	SiO ₂ ^a
Initial	332	28	533	49	–	1468	2163	1120	1506	22,000
Final	81	0.4 ^b	228	27	–	1120	1854	1429	2086	23,948
Leachate	424	32	211	8.4	10.8	9.6	1.7	0.058	0.013	0.83

^aAssuming the acid insoluble fraction is silica.^bAssuming final Cl of 10 mg/kg.

leachate, an alternate mass leached can also be derived. The results are presented in Tables 15 and 16 and Fig. 6.

Calculation of the soluble mass leached allows comparison with the soluble mass available at the start and end of the trials; estimation of initial salt loadings due to surface washing of soluble salts; estimates of remaining salt loads; and behaviour of trace metals. A further use of these assessments is a check on the accuracy of ash and leachate analyses and soluble mass calculations. Thus, it is possible to ascertain the long term potential leachate that may influence groundwater quality, if leachate was to migrate into the underlying aquifers.

5. Discussion of the geochemistry of ash leaching

The ash profile maintained a strongly oxidising environment (> 300 mV) throughout the field trial, with the only exceptions being at the start and end of the cells where the redox state was slightly oxidising (~ 45 to 160 mV). The strongly oxidising environment is considered to be related to the entry of air into the unsaturated ash profile. In the Wet Cell, the decrease in oxidising conditions in July to August 1998 is most likely due to the lower rates of air entry through the ash profile as the degree of saturation increased with the high volume of water being irrigated.

The comprehensive approach of Eighmy et al. [29] was to undertake a detailed qualitative and quantitative assessment of ash mineralogy and morphology, and thereby to compare predicted leachate chemistry using geochemical modelling with MINTQEA2 [30] and those from sequential batch leaching tests. Although they were successful in demonstrating the different mineralogical controls on leachate chemistry, they recognised the need for further mineralogical analyses to establish a more comprehensive

Table 16

Total soluble mass in leached ash and leachate: trace elements (g)

	As	B	Ba	Mo	Se
Initial	371	5909	6953	371 ^a	317 ^a
Final	193	4944	23,445	276	193
Leachate	0.74	598	3.91	9.41	75.1

^aAssuming the average from Dry Cell ash.

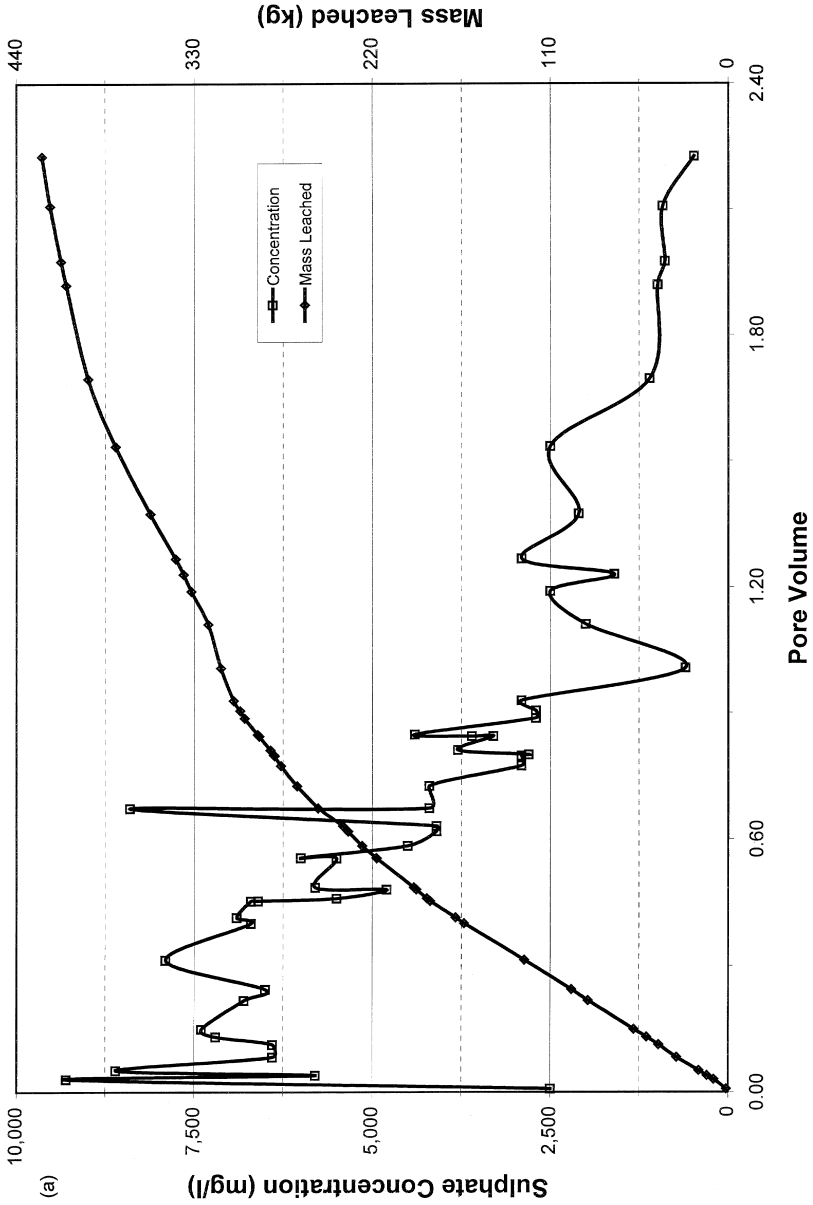


Fig. 6. Concentration and leached mass vs. pore volume (Wet Cell).

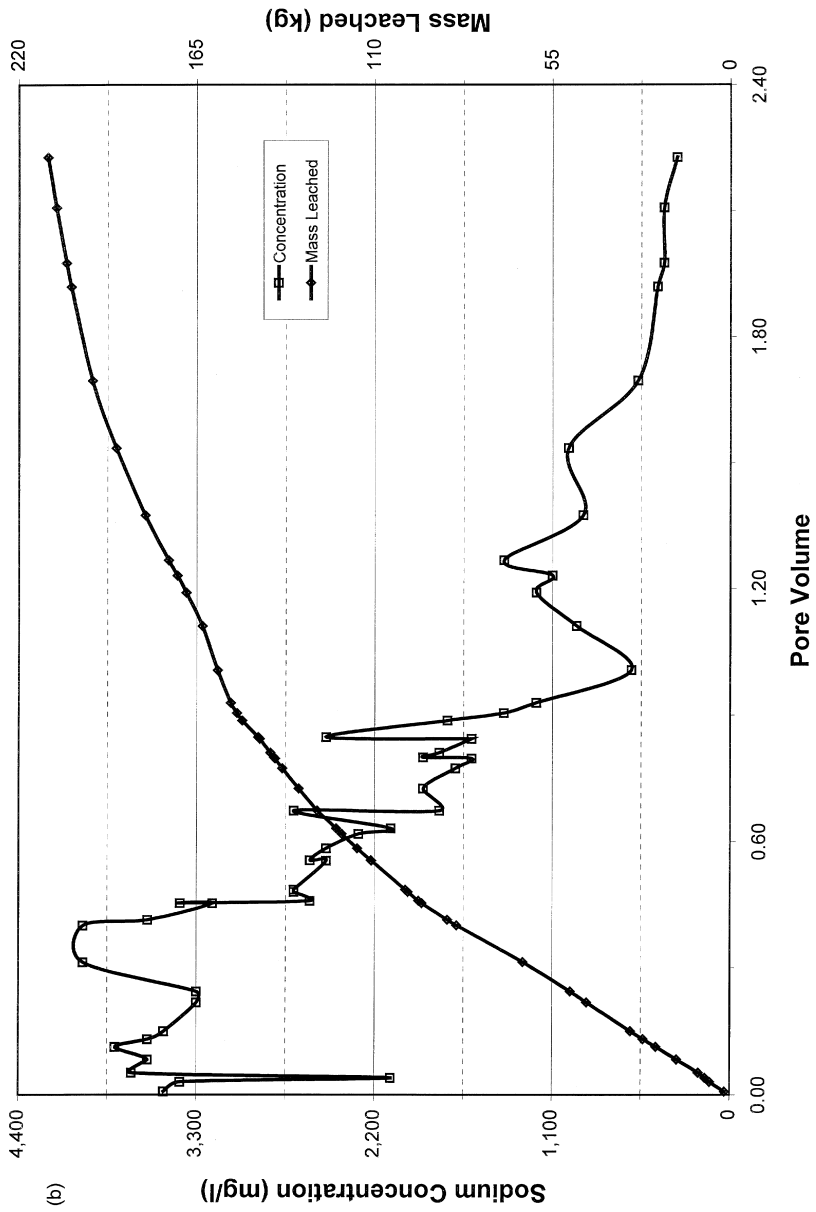


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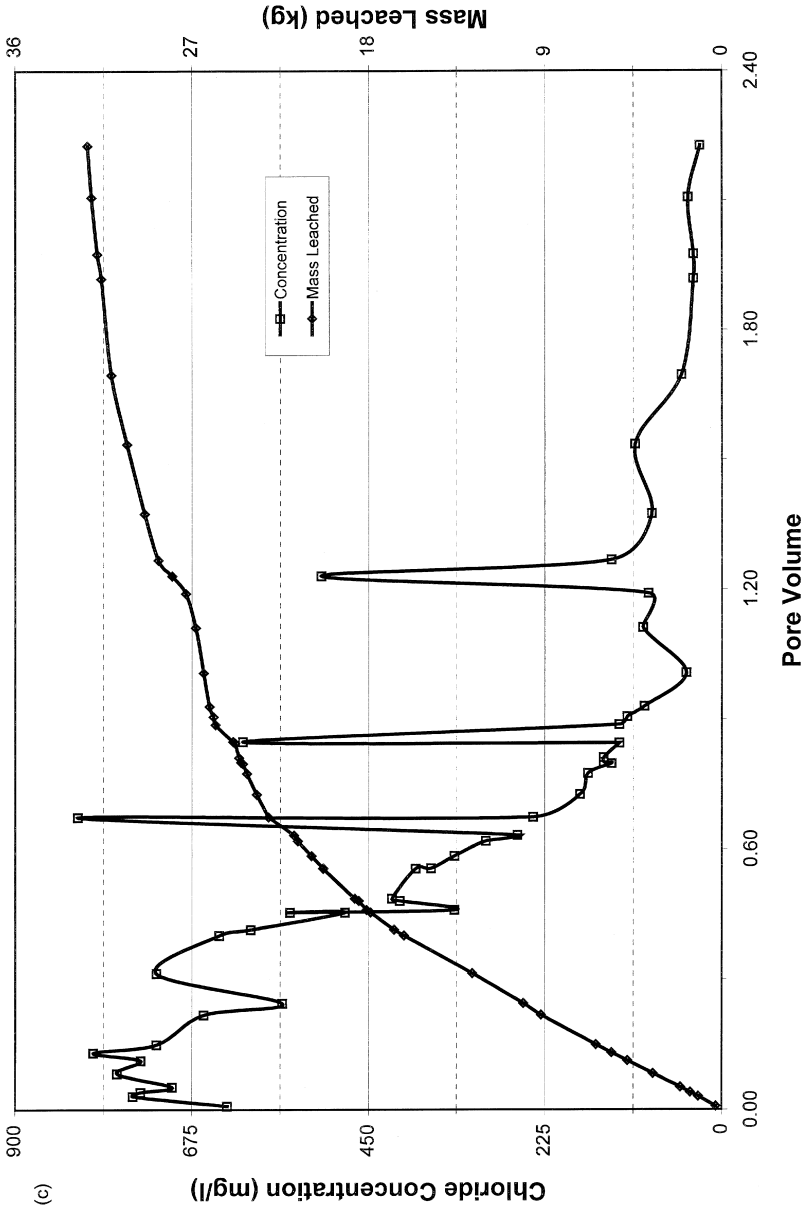


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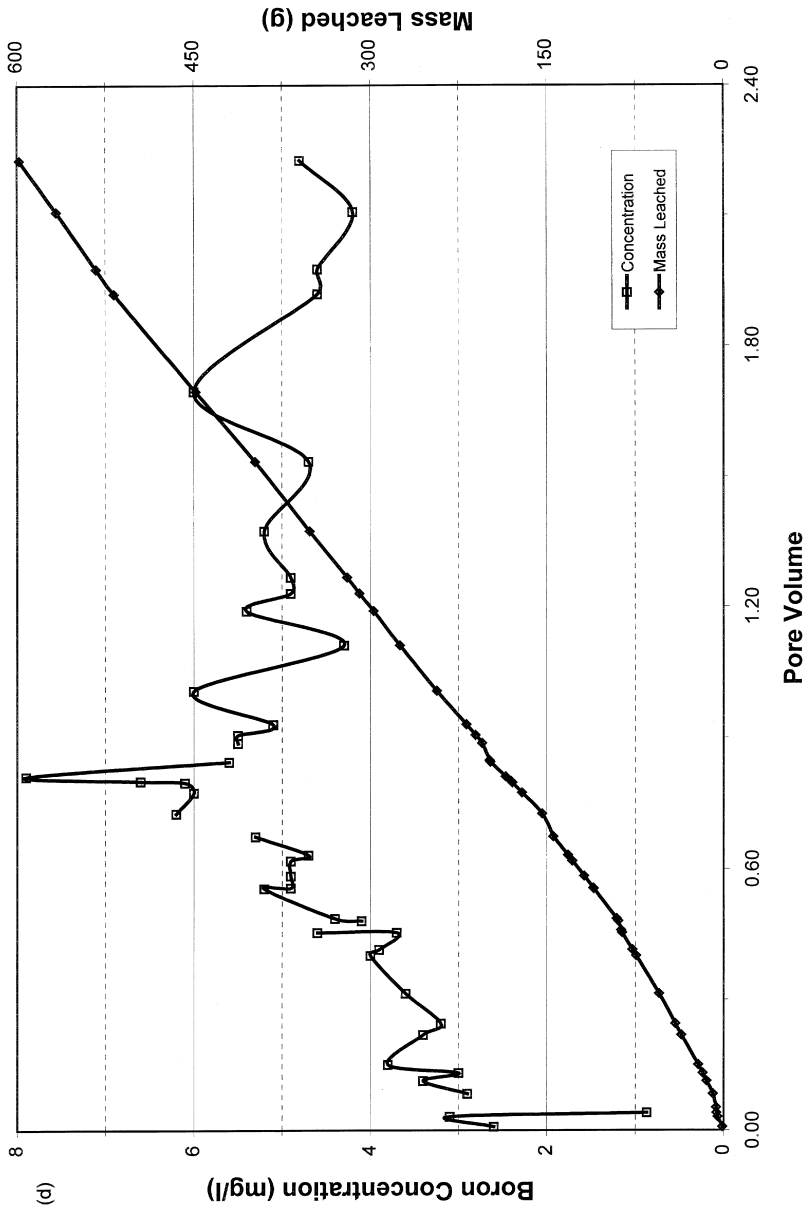


Fig. 6. (continued).

database of ash mineralogy, and therefore controls on leachate chemistry. Given the variability of ash chemistry within the Latrobe Valley, it was considered more prudent for this study [22] to analyse the resulting leachate chemistry as it might influence environmental requirements, rather than a detailed study of ash morphology.

The leachate chemistry was analysed using the PHREEQC geochemical computer model [31]. PHREEQC was used to study mineralogical associations and controls on leachate chemistry. Only the major elements and boron were modelled, given the lack of available mineral data on trace elements. The results are presented in Table 17.

The mineral saturation data indicate that the leachate is initially undersaturated with respect to sulphate minerals such as gypsum and anhydrite, although barium solubility is being controlled by the high sulphate concentrations since barite is over-saturated. The degree of saturation of barite decreases as the field trial progresses, with barite eventually becoming undersaturated in the leachate. However, with respect to carbonate species, the leachate is oversaturated with calcite and dolomite though significantly undersaturated with siderite. Although the calcite and dolomite saturation indices decrease by the end of the trial, they still remain oversaturated. The high alkalinity and exposure of the ash to atmospheric carbon dioxide diffusing through the profile would therefore be expected to lead to carbonate minerals being precipitated.

The leachate chemistry and mineral saturation data indicate that the Na, Cl, K, S and Ca are in readily soluble mineral assemblages, most likely as salts on the surface of the ash particles. The data for trace elements is less clear, although geochemical conditions such as pH, redox state and complex formation are likely to control the solubility of environmentally important elements (e.g. Se, Ba, As).

The solubility-controlling phases may also be inferred from plots of the log of the activity of particular species, such as Ca vs. SO_4 or Al vs. pH. This approach has been successfully used by several workers, including Mattigod et al. [25], Fruchter et al. [32] and Roy and Griffin [33] in analysing ash leachates from laboratory studies and field sites in the USA. The log activities of different elements and species for each leachate

Table 17

Saturation indices of Wet Cell leachate with respect to possible solubility controlling mineral phases in the ash

Mineral	Formula	Saturation Indices				
		July–Sept. 1997	Dec. 1997	Mar. 1998	June 1998	July 1998
Gypsum	$\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$	−0.49	−0.70	−0.76	−1.13	−1.79
Anhydrite	CaSO_4	−0.71	−0.92	−0.97	−1.35	−2.00
Barite	BaSO_4	0.85	0.45	−0.02 ^a	0.24	−0.22 ^a
Aragonite	CaCO_3	0.78	0.78	0.05	0.43	0.54
Calcite	CaCO_3	0.93	0.93	0.19	0.57	0.69
Dolomite	$\text{CaMg}(\text{CO}_3)_2$	1.48	1.40	−0.18	0.89	0.92
Siderite	FeCO_3	−6.58	−7.13	−5.18	–	−8.06
Witherite	BaCO_3	−3.04	−3.23	−4.38 ^a	−3.37	−3.05 ^a
Gibbsite	$\text{Al}(\text{OH})_3$	0.71	0.03	1.52	−0.47	−0.27
Ferrihydrite	$\text{Fe}(\text{OH})_3$	1.24	1.25	2.20	–	−0.14
Quartz	SiO_2	0.08	0.08	0.14	−0.03	−0.41

^aAssuming a concentration of barium at the detection limit of 10 $\mu\text{g/l}$.

sample from the Wet Cell was analysed with PHREEQC and plotted according to mineral equilibrium data, shown in Fig. 7. The various graphs presented in Fig. 7 show smooth variations in log activities, either as functions of the log activity of a particular species or of pH.

The leachate data show that Ca activities are essentially independent of pH (for the measured pH of the leachate samples, ~ 8 to 10). This suggests that mineral phases such as calcite or portlandite do not control Ca solubility in the leachate [32]. By considering the dissolution of gypsum or anhydrite, a plot of Ca^{2+} activity vs. SO_4^{2-} activity shows a linear correlation. However, if Ca^{2+} activity was being controlled by gypsum or anhydrite, as the SO_4^{2-} activity increased the Ca^{2+} activity should correspondingly decrease. If only the initial leachate data from July to October 1997 is considered (when the leachate was apparently saturated with respect to ash salts and minerals), the Ca^{2+} and SO_4^{2-} data plots according to gypsum/anhydrite solubility. As the leaching progressively removes calcium and sulphate, these species become limiting and gypsum no longer appears to control these species in the leachate.

The graph of Fig. 7b also shows similar behaviour for Na^+ activity vs. SO_4^{2-} activity. However, the initial leachate samples are several orders of magnitude below the equilibrium saturation line for thenardite (assuming a log K of -0.179 for thenardite; [31]). This suggests that despite the high solubility of thenardite, the leachate is significantly undersaturated with respect to this mineral. The cause of this is unclear, but may indicate a kinetically controlled dissolution reaction, alternate elemental distribution within the ash matrix other than a surface salt or inaccurate thermodynamic data for thenardite.

At a high pH (above 8.5), the Al data shows good correlation with crystalline gibbsite, while below this pH the graph suggests that amorphous gibbsite becomes more important in controlling solubility. This behaviour of Al is consistent with the data reported by other workers [25,32].

There were no iron speciation analyses performed on leachate samples, although the redox state and PHREEQC geochemical data suggest that the iron is predominantly in the Fe^{2+} (ferrous) state. The controls on iron solubility are less clear, however, the graph in Fig. 7d may be influenced by analytical error since most iron concentrations in the leachate were low and near detection limits (with possible interference from colloidal iron during sampling). The graph may also represent non-equilibrium conditions prevailing that limit the solubility of iron in the leachate.

The graph of silicic acid (H_4SiO_4) vs. pH indicates that silica (SiO_2) in the leachate is not controlled by amorphous silica, although, despite the high scatter of data, there may be a controlling influence of quartz. Fruchter et al. [32] demonstrated that wairakite ($\text{CaAl}_2\text{Si}_4\text{O}_{12}\cdot 2\text{H}_2\text{O}$) may be the controlling silicate mineral phase in their Montour fly ash from Pennsylvania. Roy and Griffin [33] confirmed the control of silica in leachate by mullite ($3\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$). The lack of aluminosilicate mineral speciation in Latrobe Valley ash, however, prevents further analysis of controlling silica phases in the ash.

The graph for Ba shows most samples plotting between barite and a barite–celestite co-precipitate, with no distinct trend. The data also shows that Ba concentrations are up to one order of magnitude higher than those predicted by equilibrium behaviour. This may be due to co-precipitation with strontium (giving barite–celestite or $(\text{Ba}, \text{Sr})\text{SO}_4$),

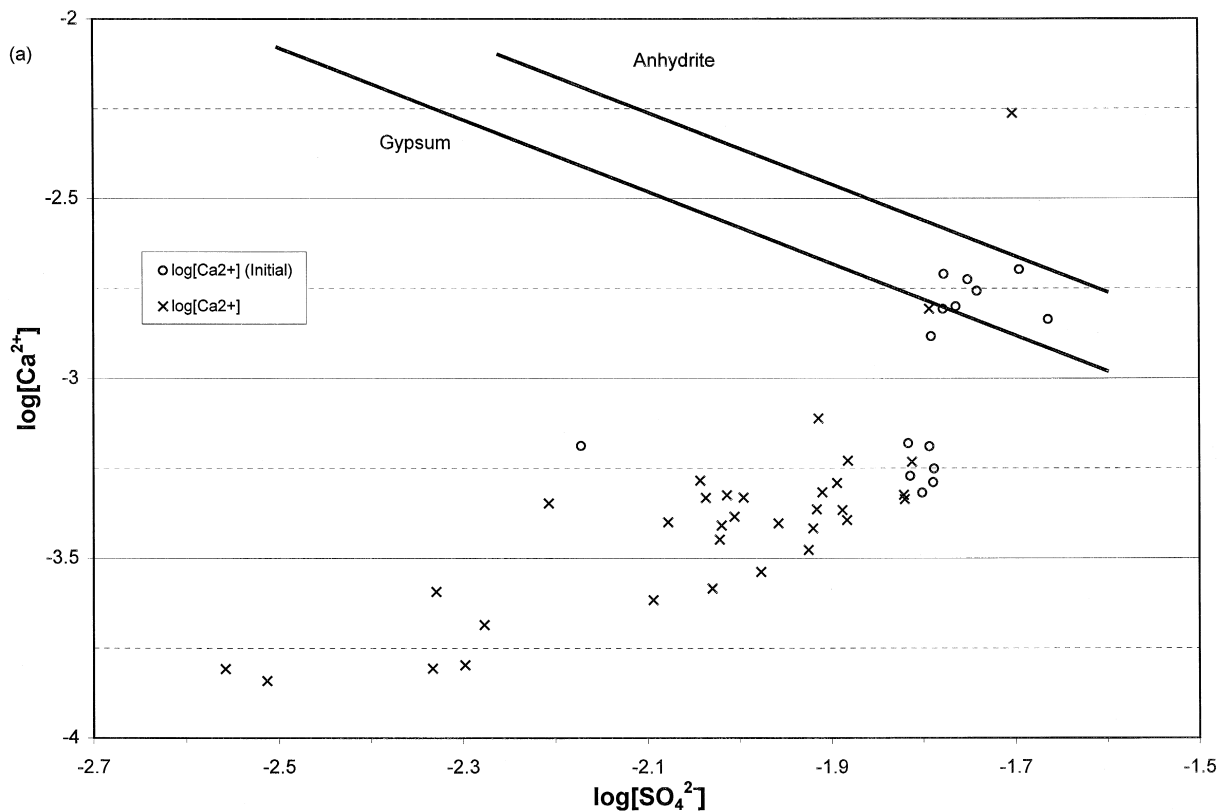


Fig. 7. Plot of PHREEQC log activity of leachate vs. mineral equilibria (Wet Cell). (a) $\log [\text{Ca}^{2+}]$ vs. $\log [\text{SO}_4^{2-}]$; (b) $\log [\text{Na}^+]$ vs. $\log [\text{SO}_4^{2-}]$; (c) $\log [\text{Al}^{3+}]$ vs. pH; (d) $\log [\text{Fe}^{3+}]$ vs. pH; (e) $\log [\text{H}_4\text{SiO}_4]$ vs. pH; (f) $\log [\text{Ba}^{2+}]$ vs. $\log [\text{SO}_4^{2-}]$. ("Initial" is from July to October 1997 — at leachate saturation).

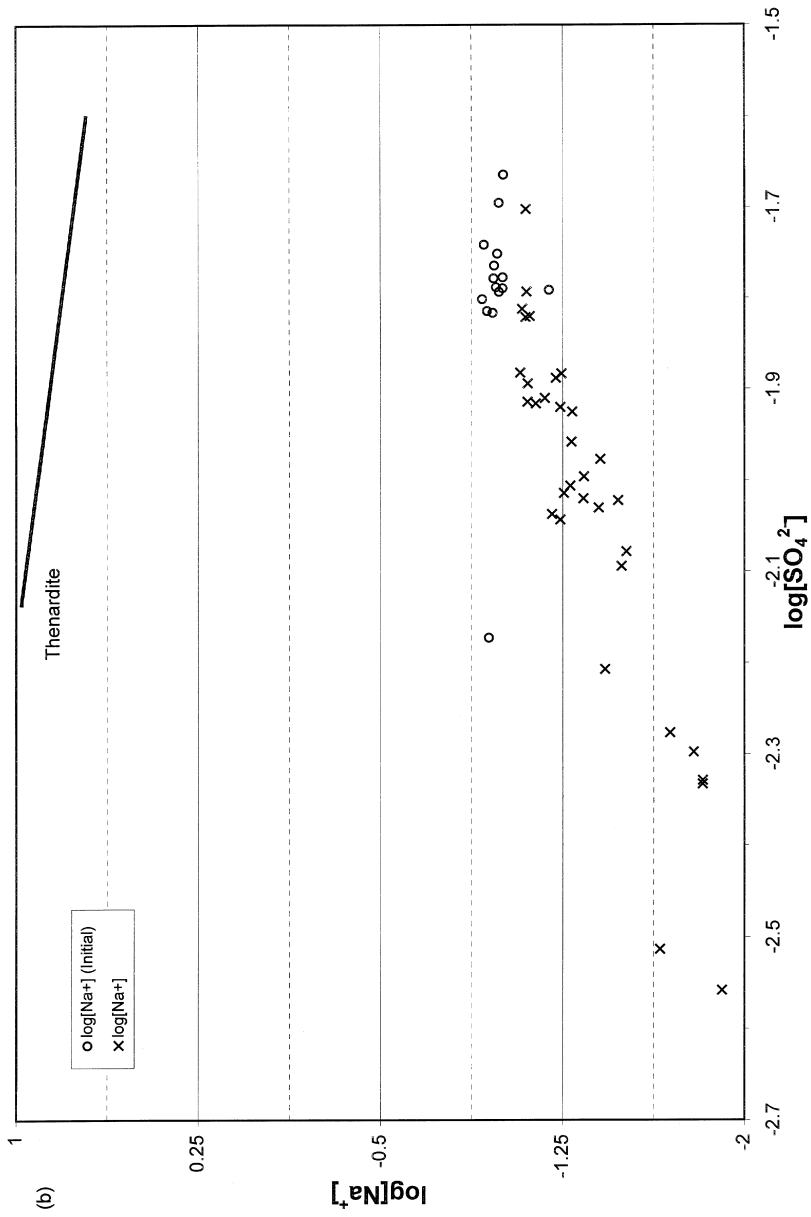


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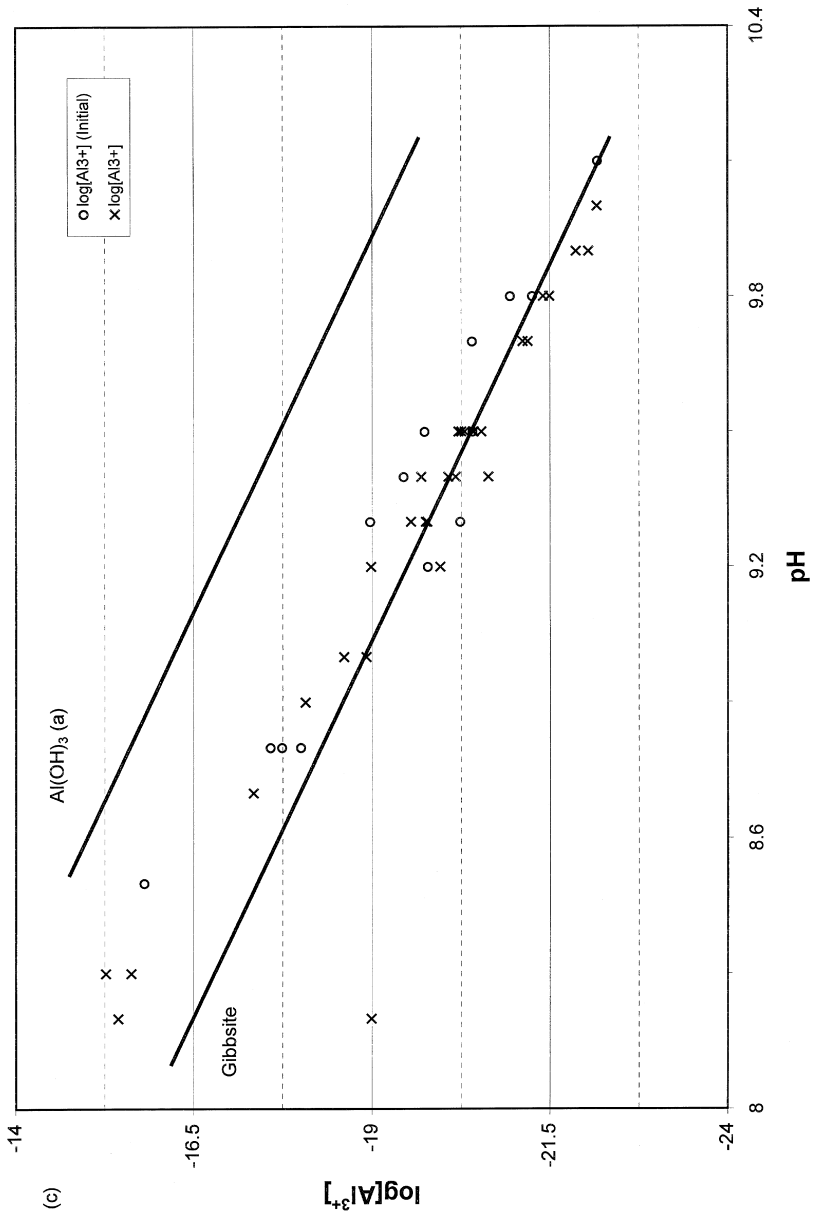


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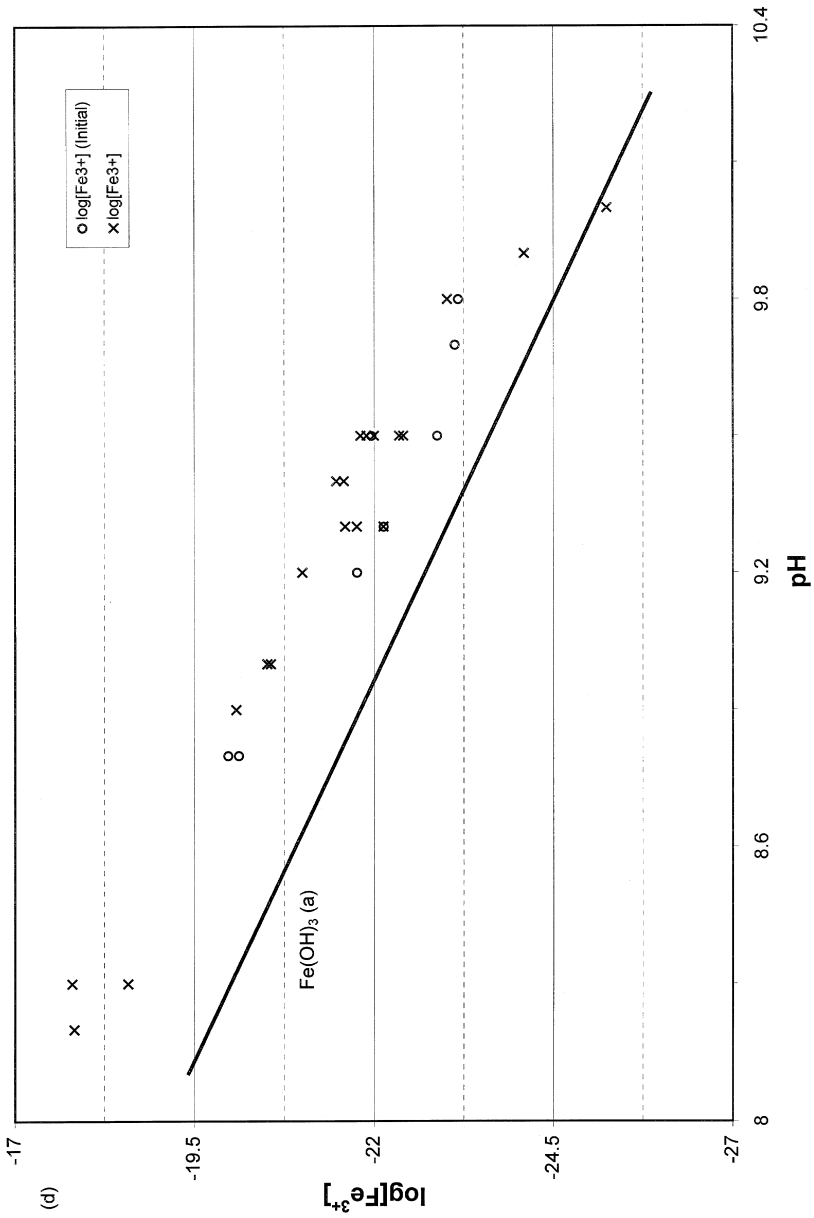


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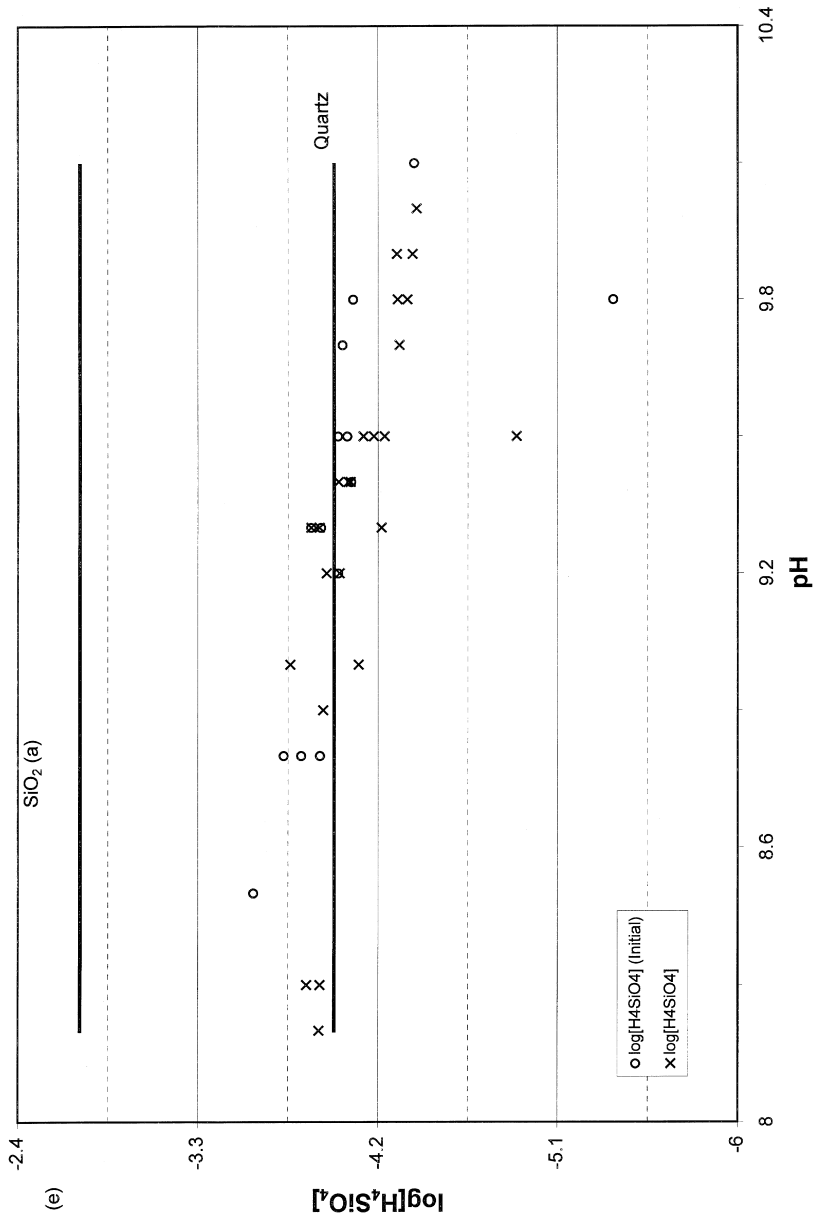


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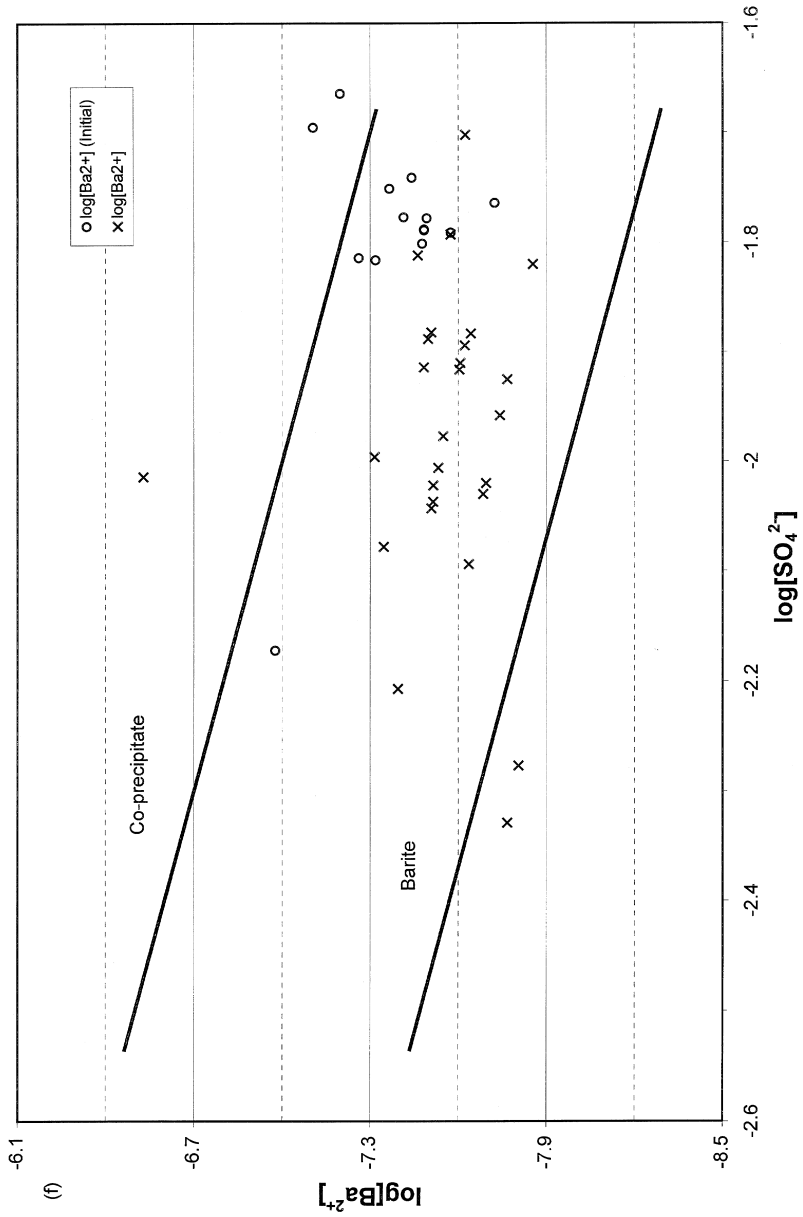


Fig. 7. (continued).

leading to an effective increase in barite solubility [32,34]. This cannot be investigated due to the absence of strontium analyses in the leachate.

The behaviour of B, although not included in Fig. 7, also warrants attention. The concentration of B gradually increased until about March 1998 (about 0.81 pore volumes), from where it slowly began decreasing, finishing at a concentration higher than the initial leachate. There is no available mineralogical speciation data for B in Latrobe Valley ashes, and other workers have failed to demonstrate that soluble borate minerals control concentrations found in ash leachates (cf. Refs. [23,27,32]). Boron is known to undergo adsorption on iron and aluminium oxide surfaces, and this may help to explain the change in boron concentrations over time [35–37]. Adsorption leads to retardation during moisture movement, thereby controlling leachate concentrations of B.

6. Conclusions

To study the management of disposal sites of leached ash, a field study was established with the construction of two HDPE-lined leaching cells. The two cells, the Dry and Wet Cells, initially showed a moderately saline and alkaline leachate. This consisted of sodium, sulphate, chloride, carbonate and calcium with minor amounts of potassium and magnesium. The only trace metals detected in minor concentrations were aluminium, arsenic, barium, boron, molybdenum and selenium.

For the Wet Cell, as the number of pore volumes of water leaching the cell increased, the salinity quickly dropped to over 90% of the initial level. The trace metal content also decreased over time, though not as significantly. The Wet Cell demonstrates that the leached ash can be more thoroughly leached with the application of about 2.4 pore volumes of water.

The mineralogical controls on leachate chemistry appear to be complex. Highly soluble salts, such as gypsum or thenardite, exert a significant control on initial leachate chemistry. The behaviour for minor and trace elements is less clear, although iron and aluminium hydroxides do appear to control their solubility. The behaviour of boron suggests a strong adsorption control, while barium behaviour suggests co-precipitation with strontium controls these species.

The field studies have indicated that placement of leached ash excavated from an ash pond can meet environmental criteria, although the trade-off is an initial moderately saline leachate. However, studies of the physical behaviour of leached ash have demonstrated that leachate generation rates at disposal sites within an overburden dump can be minimised due to the high porosity, moisture storage capacity and unsaturated behaviour of the ash. It may be possible to further engineer the placement of the ash to get the maximum benefit from the hydraulic characteristics of ash in minimising generation of leachate.

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