Use of phytocaps in remediation of closed landfills - correct selection of soil materials

I.R. Phillips, M. Greenway and S. Robertson

Abstract

Phytocapping is an emerging technology gaining interest as an alternate capping system in the management of closed landfills, particularly where leachate production occurs primarily through groundwater intrusion rather than surface infiltration. A field trial was undertaken at a former putrescible landfill to compare the performance of a phytocap and an engineered cap. The engineered cap comprised compacted subsoil, overlain by a gravel drainage layer, overlain by sandy clay loam topsoil. The phytocap comprised a layer of topsoil only. A range of native trees, shrubs and grasses were planted into each cap, and irrigated daily with landfill leachate of varying strength (0, 40 or 100%). Laboratory and field-testing evaluated the effect of leachate on soil physical and chemical properties. Deterioration in the topsoil's hydraulic properties impaired leachate infiltration and encouraged surface erosion for both systems. Laboratory studies found that aggregate instability due to slumping was the major cause for the decline in soil hydraulic properties. The topsoil sorbed appreciable amounts of added ammonium, organic nitrogen and organic carbon, and transformed much of the added ammonium to nitrate within a 14-day period. Thus, the capping system has the potential to remove leachate contaminants if the hydraulic properties of the topsoil can be improved. The trial has shown the importance of correct testing prior to selecting soil materials for use in capping systems where infiltration of leachate and rainwater is encouraged rather than discouraged.

Key words: nitrogen, carbon, phytocap, phytoremediation, landfill leachate, capping systems

INTRODUCTION

Leachate management continues to be one of the major issues facing regulatory authorities responsible for the remediation of closed landfills. Various strategies for minimising leachate release into the environment can be engineered into landfill design such as drainage collection systems, cut-off walls, reactive permeable barriers, collection and recirculation systems, and the construction of low-permeability (engineered) surface

Received June 2004; revised and accepted August 2004

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caps (Smith et al. 2003; Henken-Mellies and Gartung 2004). Engineered caps have been widely used as an effective method to minimise water entry into landfills, and consequently, the potential for leachate generation (Henken-Mellies and Gartung 2004). Consequently, engineered capping systems are recommended in environments where infiltration (rainfall, snow melt, etc.) exceeds evapotranspiration.

In sub-tropical environments such as that experienced in Brisbane, Australia, leachate generation through infiltration of rainwater is of much less importance than that generated through groundwater inflows. Of the 150 former landfills within the Brisbane area, most are in direct contact with groundwater, and >80% of the leachate generated results from groundwater intrusion. Furthermore, Brisbane's rainfall is summerdominated and can occur from high-intensity storms, which encourage surface runoff rather than infiltration. Field research by Brisbane City Council (Thorley and Boczek 2001) indicated that infiltration in Brisbane's

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sub-tropical environment can be limited to less than 0.05% of precipitation without the use of multilayered caps, and even a 400 mm uncompacted subsoil layer reduced infiltration to <2%. The incorporation of fully engineered caps in landfill management at many Brisbane closed landfills may therefore not be warranted.

Recently, less-costly capping designs that incorporate leachate-tolerant plants have been investigated as an alternative capping system to engineered caps. These alternative designs involve planting trees, shrubs and grasses directly into the landfill cover material, or into a thin layer of topsoil (approximately 300 mm) above the landfill cover, and have been termed phytoremediation caps (or phytocaps). The phytocap principally involves the use of plants that are tolerant to highly saline, ammonium-rich leachate, and is closely related to the established remediation technology referred to as phytoremediation (Brooks 1998). The plants perform a variety of tasks, which include creating a soil-water deficit through transpiration, encouraging soil microbial activity for removing (within the soil biomass) and converting leachate chemicals (e.g. organic nitrogen) into usable forms (inorganic nitrogen), acting as a sink for leachate contaminants through plant uptake, and improving soil structure within the capping system.

In 2002, Brisbane City Council (BCC) commenced a major innovative project to investigate the applicability of phytocapping as a method of managing leachate at a former major landfill. This represents one of the first phytoremediation trials to be undertaken at a Queensland landfill, and possibly the first of its kind in Australia (Phillips 2003). The principal aims of the trial were to:

- assess whether the concept of phytoremediation can be incorporated into landfill capping design;
- identify a range of native plants that have the potential to treat leachate on-site; and
- to evaluate the suitability of the phytocap as an alternative capping system to the more costly engineered cap.

The work reported in this paper identifies some of the major issues associated with selection of capping materials, and provides an overview of the cap performance in comparison with an engineered cap. The suitability of various plant species used in the trial to tolerate landfill leachate will be presented by Greenway *et al.* (2004).

MATERIALS AND METHODS

Site description

The field trial was undertaken at the former Nudgee Landfill site (27°21.20'S 153°055'E), in Brisbane, Queensland, Australia. The landfill is underlain by 1.4-4.0 m of alluvial floodplain sediments (clay, silt and gravel), which in turn are underlain by extremely weathered basalt. Groundwater at the site is essentially tidal, and the dominant flow direction is to the north in response to the general surface drainage direction. The former Nudgee Landfill has a tipping history extending from September 1976 to January 1988 (136 months), and was used as a general refuse tip over this period. It is currently operating as a hard fill site, and an on-site transfer station has been in operation since 1993. The site covers an approximate area of 720 000 m², and extends to an approximate depth of 20 m. Initial construction of the landfill did not include a liner system, and a significant proportion of the waste mass is in direct contact with the tidal groundwater (BCC, personal communication).

Description of the field trial

The trial comprised twelve plots (22.5 m long by 12 m wide), and included two capping designs (engineered and phytocap), three leachate qualities (0, 40 and 100%) and two replicates of each plot (replicates 1 and 2). The engineered cap comprised a compacted clay layer (about 500 mm thick) above the existing landfill cover, underlying a gravel drain layer (300 mm thick), underlain by sandy clay loam topsoil (300 mm thick). A geotextile layer was placed between the gravel and topsoil layers to minimise the movement of fine materials into the gravel matrix. Leachate was sourced directly from the landfill, and its general composition is presented in Table 1. Leachate was mixed with proportional amounts of reticulated water to obtain solution concentrations of 0% (control), 40% and 100% leachate strength.

Preliminary studies (Ashwath and Hood 2001) identified 18 plant species that were suitable for the trial, which included a number of trees (acacias, eucalypts and grevilleas) and grasses (*Vetiva* and *Juncus*). Species were planted in a randomised block design, and drip irrigated at a daily rate of 3.7 mm (approximately equivalent to 1.5 times the annual rainfall). Plant characteristics such as height and girth were monitored throughout the trial, and leaf samples of each plant were collected for total N and total C analysis using mass spectrometry. Soil from across each plot was analysed for gravimetric water content, pH, EC, and 2M KCl NH₄ and NO₃, using standard techniques.

Parameter	Value
EC (μS/cm)	15 000
рН	7.1
Total organic carbon (TOC)	230
BOD ₅	21
Total Kjeldahl nitrogen (TKN)	510
NH ₄ –N	330
NO ₃ –N	4.9
Total alkalinity (CaCO ₃ equivalent)	1700
SO ₄	240
СІ	5000
Са	130
Mg	180
Na	2900

Table 1. Composition of leachate from the Nudgee Landfill (all units in mg/L unless otherwise stated)

Table 2. Relevant physical and chemical properties of the sandy clay loam topsoil

Parameter	Topsoil		
Texture	Sandy clay loam		
Stones >20 mm	2–3%		
Large particles >2 mm	10%		
рН	7.5		
EC (µS/cm)	260		
Organic carbon (%)	1.22		
Oxalate-Fe (%)	0.38		
Oxalate-Al (%)	0.13		
CI (mg/kg)	273		
NO ₃ (mg/kg)	1.5		
P (mg/kg)	99		
SO ₄ (mg/kg)	33		
Ca (cmol _c /kg)	4.42		
Mg (cmol _c /kg)	2.93		
K (cmol _c /kg)	0.66		
Na (cmol _c /kg)	1.16		
Cu (mg/kg)	3		
Zn (mg/kg)	3.58		
Coarse sand (%)	39		
Fine sand (%)	40		
Silt (%)	6		
Clay (%)	15		

Hydraulic conductivity and aggregate stability

Soil used in construction of the surface layer (a sandy clay loam) was studied because this material was in direct contact with the leachate. Relevant initial properties of this soil are presented in Table 2.

The soil's saturated hydraulic conductivity (k_{sat}) was determined using tapwater (sodium adsorption

ratio (SAR) = 1, 40% leachate (SAR = 12) and 100% leachate (SAR = 27) to reflect treatments used in the field trial. Approximately 150 grams of soil were packed into a glass burette, and a constant depth of solution of about 5 cm was maintained above the soil surface. The soil was sequentially leached as stated in Table 3. Solution was continuously added to each soil column until the discharge rate (i.e. volume of outflow per unit time) remained constant for three successive readings. Alternating the solutions between leachate and tapwater simulated conditions expected in the field experiment where leachate-affected soil will be intermittently leached with low ionic strength rainwater. The 0.1 M CaCl₂ solution was used to determine if any loss of k_{sat} due to leachate and/or tapwater could be recovered using a concentrated solution containing a multivalent cation. The k_{sat} was calculated as:

$$q = (V \div At) = -k_{\text{sat}} \times dH/dz \tag{1}$$

where

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q =	=	the Darcian	flux	density	(m ²	$^{2}/m^{2}$	² .s);
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- V =volume of drainage (m³);
- A = cross-sectional area available for flow(m²);

$$=$$
 time (s);

- k_{sat} = saturated hydraulic conductivity (m/s);
- H = hydraulic head (m);
- z = distance (m); and
- dH/dz = the hydraulic gradient (m/m); the negative sign indicates water flows in the direction of decreasing *H*.

Aggregate stability testing was conducted (n = 2)using undisturbed samples of sandy clay loam topsoil material from the field trial. Aggregates were immersed in solutions of deionised water, tapwater, 40% leachate and 100% leachate, and the extent of slaking and dispersion was evaluated visually. Individual jars (8×250 mL) were filled with solution to attain a ponded depth of 5 cm. A soil aggregate approximately 1 cm in diameter was then carefully placed in the solution. The aggregate was visually observed for ten minutes, and the effects of solution type on aggregate stability were recorded. The aggregate was left undisturbed in the jar for 20 hours, after which its condition was recorded. Observations of the effect of solution on aggregate stability involved the presence of air escaping from the aggregate seen as bubbles in the solution, slumping of the aggregate, and clarity or cloudiness of the solution from dispersed fine material.

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Sequence	Sequence of solutions added						
1	Tapwater	\rightarrow	Tapwater	\rightarrow	Tapwater	\rightarrow	0.1 M CaCl ₂
2	40% Leachate	\rightarrow	Tapwater	\rightarrow	40% Leachate	\rightarrow	0.1 M CaCl ₂
3	100% Leachate	\rightarrow	Tapwater	\rightarrow	100% Leachate	\rightarrow	0.1 M CaCl ₂

Table 3. Leaching sequence to assess solution type on k_{sat}

Sorption isotherms and transformation

Sorption isotherms (n = 2) were determined for the sandy clay loam topsoil using the standard batch technique. Leachate was diluted with deionised water to obtain eight (8) initial solutions, and these solutions represented 0, 5, 10, 20, 40, 60, 80 and 100% leachate. The range of total dissolved N (TN), NH₄-N, total dissolved carbon (TC), total dissolved organic carbon (TOC) of these solutions were: 0-500, 0-430, 0-600, and 0-380 mg/L, respectively. Soil (5 g on an equivalent air-dry basis) was weighed into a series of 50 ml polypropylene centrifuge tubes, and 25 mL of solution added. The suspension was shaken end-over-end for two hours, then centrifuged at 3000 rpm for 30 minutes, and passed through a 0.45 µm filter prior to analysis. The initial and final solutions were analysed for NH₄ (flow injection analysis techniques (FIA)), and total N, TC and TOC (Shimadzu Total Carbon and Nitrogen Analyser). The difference between total N and NH₄ concentrations was assumed to represent the total dissolved organic N (TON) fraction. Ammonium, TON and TOC adsorption data were fitted to the Freundlich equation:

$$S = k_{\rm f} c^n \tag{2}$$

where

- S = the increase in sorbed NH₄ (TON or TOC) per unit weight of soil (mg/kg);
- c = the final NH₄ (TON or TOC) solution concentration (mg/L);

 $k_{\rm f}$ and *n* are empirical fitting constants

Nitrogen transformations in the sandy clay loam topsoil were investigated using deionised water, tapwater, 40% leachate and 100% leachate. Approximately 15 g of soil (air-dry, <2 mm) were placed into individual 20 mL polypropylene syringe barrels (Chux cloth and GF/A filter paper in base of syringe; Chux cloth on soil surface) and leached with approximately 15 mL of one of the above solutions. The syringe barrels were covered with perforated plastic film, and placed in a vertical position. A sample of the soil solution was extracted after 0, 1, 3, 7 and 14 days by highspeed centrifugation (10 000 rpm for 1 hour at 4°C) using the procedure by Elkhatib et al. (1987). The extracted solutions were analysed for NH₄ and NO₃ (FIA), and total N, TC and TOC (Shimadzu Total Carbon and Nitrogen Analyser). The production of NO₃ was modelled using the average (n = 2) NH₄ and NO₃ data, using:

$$c_{\rm NO3} = a \times t^{b1} \times c_{\rm NH4}^{b2} \tag{3}$$

where

- c_{NO3} = concentration of NO₃ in the extracted solution (mg/L);
- $c_{\rm NH4}$ = concentration of NH₄ in the extracted solution (mg/L);

t is time (days); and

a, b1 and b2 are constants.

Equation (3) allows the effects of time and solution NH_4 concentrations to be included in NO_3 production, and has been successfully used to model P kinetics in soils (Gerritse 1993).







FIGURE 2. DETERIORATION IN PHYSICAL PROPERTIES OF THE FIELD PLOTS DURING EARLY STAGES OF THE TRIAL, DUE TO (A) SEDIMENT TRANSPORT, AND (B) EROSION

RESULTS AND DISCUSSION

Aggregate stability

All aggregates slumped immediately on immersion in the various solutions (Figure 1). Air was observed to diffuse out of the aggregate into the solution, which brought about disintegration of the aggregate. Slumping can be qualitatively assessed based on a score of 0 to 4, where 0 represents no slumping, 1 represents slumping largely around the aggregate edges, 2 represents aggregate collapse into angular pieces, 3 represents aggregate collapse into small (<2 mm diameter) rounded pieces, forming a cone shape, and 4 represents aggregate collapse into single grains. A score of 3 best described the aggregate stability for the sandy clay loam topsoil (Figure 1), and is consistent with the deterioration in soil physical properties observed in the field trial (Figure 2). The tendency of the topsoil to slump on immersion can be attributed to its lack of binding materials such as organic matter and sesquioxides (Table 2).

Dispersion was only observed in the deionised water treatment (Figure 1(a)), and recorded a dispersion index (on a rating between 0 to 4) of about 2 (i.e. 'moderate dispersion with obvious milkiness'). The absence of this effect in the other treatments may be attributed to the higher ionic strength of the tapwater and leachate solutions, which can reduce dispersion through compression of the diffuse double layer. When >6% of the cation exchange capacity of a soil is occu-



FIGURE 3. EFFECT OF LEACHING SOLUTION ON K_{SAT} FOR THE SANDY CLAY LOAM TOPSOIL



FIGURE 4. (A) AMMONIUM (NH4) AND (B) ORGANIC N SORPTION ISOTHERMS FOR THE SANDY CLAY LOAM TOPSOIL

pied by sodium (Na), soils are regarded as sodic. On wetting, the Na cannot bind the clay particles together, and since Na cannot overcome the repulsive forces, clay dispersion occurs. However, increases in solution concentration can overcome this repulsion, thereby causing reducing dispersion. The topsoil at the Nudgee trial had an exchangeable sodium percentage (ESP) of 12%, hence it was sodic. Re-wetting with deionised water (EC = 1.3 μ S/cm) would encourage dispersion, as observed in Figure 1. However, the concentration of tapwater (EC = 394 μ S/cm; which most likely also contained the divalent cations Ca and Mg), and leachate (EC_{40%} = 8030 μ S/cm and EC_{100%} = 18 200 μ S/cm)



FIGURE 5. ORGANIC CARBON SORPTION ISOTHERM FOR THE SANDY CLAY LOAM TOPSOIL

appeared to be sufficiently high to overcome the repulsive forces, thereby avoiding clay dispersion.

Effect of leachate on saturated hydraulic conductivity (k_{sat})

The effect of leaching solution on k_{sat} for each soil material is presented in Figure 3. Initially, k_{sat} was about 2×10^{-5} m/s, irrespective of leaching solution (Figure 3). Subsequent leaching with tapwater reduced k_{sat} , particularly for those soils initially leached with leachate. For these treatments, tapwater reduced k_{sat} to about 1×10^{-6} m/s, and subsequent addition of leachate or 0.1 M CaCl₂ failed to increase k_{sat} to its initial value. This suggests that the detrimental effects of sequential application of leachate and tapwater on the hydraulic properties of this sandy clay loam soil were irreversible (Scotter 1985).

The reductions in k_{sat} may be a result of mechanisms such as slumping, swelling pressures, and/or clay dispersion. Of these mechanisms, swelling pressures and dispersion were considered to be of minor importance. Firstly, the soil contained a relatively low clay percentage (Table 2), and, although not quantified, the clay mineralogy is unlikely to contain highly reactive materials such as smectite. Secondly, earlier data showed that little dispersion of the clay-sized material occurred in the presence of tapwater and leachate (Figure 1). Thus, it was concluded that slumping represented the dominant mechanism for the deterioration in aggregate stability.

During the k_{sat} experiments, it was observed that the surface few millimetres of the soil columns contained a

high proportion of very fine soil material (probably generated through slumping), and this material may have formed a low-permeability layer that inhibited infiltration of applied solutions. The finding that both the leachate and the 0.1 M CaCl₂ could not reflocculate this material, suggests that the formation of a low-permeability layer was an irreversible process. Therefore, blockage of conducting pores by a layer of colloidal material after slaking (particularly at the soil surface) may be the most likely cause for the large, permanent decrease in k_{sat} observed in these soils. A similar finding was reported by Minhas and Sharma (1986) for a clay loam soil.

Nitrogen and carbon adsorption isotherms

The sorption isotherms clearly show that the topsoil has a good ability to remove leachate NH_4 (Figure 4a). Cation exchange was considered to be the primary removal mechanism, with leachate NH_4 undergoing exchange with resident exchangeable Ca, Mg and Na (Table 2). The data were very well described by the Freundlich Equation, with correlation coefficients of around 0.99 (Figure 4a).

The data also showed that the topsoil removed significant amounts of organic N, and that the maximum increase in adsorbed concentration was about 100 mg/ kg (Figure 4b). Organic compounds which contain N (e.g. amino acids, polypeptides, and proteins) can be sorbed by various processes, such as: hydrophobic sorption, hydrogen bonding, ligand exchange, cation exchange and anion exchange (Thurman 1985; Tisdale *et al.* 1993). The short duration of the isotherm study,



FIGURE 6. (A) EFFECTS OF TIME AND SOLUTION TYPE ON WATER-SOLUBLE NH_4 AND NO_3 CONCENTRATIONS IN THE SANDY CLAY LOAM TOPSOIL, AND (B) PREDICTED SOLUBLE NO_3 CONCENTRATIONS

coupled with no obvious increases in inorganic N, suggests that the loss of organic N did not result from microbial activity. It is possible that this loss was due to reactions with the soil charged-components (i.e. clay minerals, organic matter, and/or Fe oxides) since decreases in soluble organic carbon were also found to occur (Figure 5). Kaiser and Zech (2000) and Kaiser (2001) studied the sorption of dissolved organic carbon, nitrogen, phosphorus and sulphur by a range of soil materials. These workers found that dissolved organic N sorbed to amorphous Fe and Al hydrous oxides almost completely, and that no sorption maxima were reached. Sorption by clay minerals such as kaolinite and illite, however, was found to be non-linear, and a maximum sorption value was observed. This type of isotherm was also observed for the topsoil (Figure 4b). This trend was not observed for dissolved organic carbon (Figure 5). This is consistent with the observations of other workers (Kaiser and Zech 2000) who found sorptive retention of organic N to be less than that of organic C. This relationship is also supported by the smaller value of *n* in the Freundlich equation for organic N (n = 0.44) relative to that for organic C (n = 0.79). Reduced sorption of organic N relative to organic C has been attributed to much of the N-containing compounds being present in the weakly sorbed hydrophilic fraction of dissolved organic matter (Jones and Hodge 1999; Kaiser and Zech 2000).

Nitrogen transformations

Solution NH_4 concentrations decreased as a function of time in all treatments, but these effects were most pronounced in the 40% and 100% leachate solutions (Figure 6a). During the early time period (<1 day), the reduction in solution NH_4 levels can be largely attributed to cation exchange with resident cations, as evidenced by the adsorption isotherms (Figure 4a).

Solution NH₄ concentrations continued to decline throughout the 14-day study (Figure 6a). Ammonium sorption is commonly regarded as *instantaneous*, so mechanisms additional to cation exchange were contributing to the loss of solution NH₄. Figure 6a clearly shows that solution NO₃–N concentrations steadily increased between day 3 and day 14, particularly during the later stages of the experiment. It is well known that, under appropriate conditions, solution NH₄ is readily transformed to NO₃–N (Tisdale *et al.* 1993). The lag-time of about two days between NH₄ addition and NO₃ production can most likely be attributed to the time required for establishment and growth of the nitrifying population (Tisdale *et al.* 1993).

The nitrification rate for soil treated with 40% leachate varied between 0.5 and 2.4 mg/kg/day, and between 1.2 and 8.5 mg/kg/day for the 100% leachate treatment. These rates were consistent with those reported in the literature (Tisdale *et al.* 1993; Phillips 2002a). This suggests that nitrification was a major cause for the decline in solution NH₄, and that during prolonged incubation (i.e. between irrigation events), a significant proportion of solution NH₄ not retained by the soil exchange sites could be transformed to NO₃, and subsequently lost through leaching back into the strong reducing (Eh < -100 mV) environment of the landfill and/or plant uptake (Phillips 2002b).

The rate of NO₃ production (Figure 6b) was simulated very well by Equation (3), and values for the parameters a, b1 and b2 for the 40% and 100% leachate treatments are presented in Table 4. Since N dynamics

are not typically described using this relationship, the values of the parameters cannot be compared with published literature; however, it would be relatively straightforward to incorporate this equation into existing solute transport models to describe soil N coupled leaching–transformation behaviour in the design of capping systems.

Table 4. Values of the parameters obtained by fitting	
Equation (3) to the NO ₃ production data, and associate	d
correlation coefficient (r ²)	

Solution	а	b1	b2	r ²
40%	4.82	-0.44	-3.36	0.98
100%	9.19	0.19	-4.23	0.90

IMPLICATIONS FOR CAPPING DESIGN

The physical, chemical and biological properties of soil materials used in the construction of capping systems need to be fully understood before appropriate selection of materials can be made. In situations where infiltration is to be minimised, clay soils that exhibit a very low permeability and a resistance to desiccation would be the favoured material. However, for phytocaps in which infiltration of surface-applied leachate is required, a soil material that can maintain an open network of pores would be the preferred choice. Importantly, the porous structure of the final design must maintain pore connectivity between the cap surface and the bulk soil matrix, because, if infiltration rates are adversely affected through slumping, dispersion, and/ or compaction, then the system will fail. In the field trial reported here, failure of the capping system resulted in limited infiltration of applied leachate, and considerable erosion of the cap surface. Remediation strategies have, however, been put in place to rectify this problem at the Nudgee Landfill, and involved spreading a 50-75 mm depth of mulch across the surface of the topsoil. This has been observed to reduce runoff velocities, and hence erodability, and to encourage a more even spreading of the applied leachate across the plot.

The topsoil did have good chemical and biological properties, as expressed in its ability to sorb added NH_4 , organic N and organic C. These properties play an integral role in removing leachate contaminants from the solution phase, thereby reducing environmental impacts, and in transforming contaminants into forms favourable for plant uptake.

The findings from this study have provided excellent baseline information for developing appropriate designs for phytocap systems, and have emphasised the importance of correct selection of materials for use in their construction.

ACKNOWLEDGEMENTS

We wish to thank Brisbane City Council for funding support and for making a field site available for undertaking this project. We also acknowledge the work of Dr Nanjappa Aswhath from Central Queensland University, and Mr Richard Yeates from Phytolink Australia, during the early stages of the trial.

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