Sources and Fluxes of Water and Salt Below a Regional Groundwater Discharge Complex, South-Eastern Australia.

submitted by
Nikki Michelle Howes, B.Sc. (Hons.)

as a requirement in full for the degree of
Doctor of Philosophy
in the
School of Chemistry, Physics and Earth Sciences
Flinders University of South Australia

February 2003

Table of Contents

Abstract

Regional groundwater discharge zones accumulate large quantities of salt as hypersaline subsurface brines in arid and semi-arid environments. However, relatively little is known about the links between hydraulic and hydrochemical processes affecting water and solutes below the large complexes of playa lakes that are common throughout Australia and many other parts of the world. This thesis has investigated, using a combination of hydraulic, hydrochemical and isotopic methods, variability in hydrochemical and hydraulic processes at a range of scales (10^{-2}–10^{3} m) in the Raak Plain groundwater discharge complex of south-eastern Australia.

The study found that subsurface brine compositions below the playas at Raak Plain are essentially evapo-concentrated versions of the seawater-like inflow, despite variations in physical playa characteristics such as size, shape, and the presence / absence of surface water bodies and salt crusts. Small differences in ionic ratios in the top 5 cm of sediment pore waters occur due to seasonal precipitation and dissolution of surface salt efflorescences, and fractionation due to different rates of diffusive transport of ions across steep concentration gradients. Chemical and isotopic data and hydraulic modelling suggest that variability in playa physical characteristics is due to different vertical hydraulic gradients across a 30 m thick aquitard separating the regional aquifer from the local dune recharge areas. This results in locally recharged groundwater either flowing directly onto the playa surface, forming surface water bodies and salt crusts, or circulating through the deeper aquifer system before being discharged. The hydraulic modelling, in particular, highlighted the delicate balance between local and regional flow systems in groundwater discharge complexes.

Despite the importance of local flow systems, and separation of the playas from the regional aquifer by a 30 m thick aquitard, ^{36}Cl data and playa-scale solute balances suggested that regional groundwater is still the major source of solutes to the playas at Raak Plain, with groundwater and solute residence times in the discharge complex of less than 50 k yrs, and about 2 k yrs required for accumulation of the present brines. Such short accumulation times support brine leakage by some mechanism, and increases in regional groundwater salinity below Raak Plain also appear to be due to mixing with brine. However, [Cl^-] vs depth profiles below the playas suggest that the subsurface brine bodies are currently confined within the top 10 m of the aquitard, and density-driven convection below the playas has been ruled out due to the strong upward hydraulic gradients there. One mechanism for brine leakage, suggested by the hydraulic modelling, is via lateral movement in the aquitard along sandy layers into the local recharge zones where downward hydraulic gradients allow advection or diffusion into the regional aquifer. Time scales for salinization by this process would be of the order of 10^3 to 10^6 yrs.

The possibiility of brine leakage over the history of the discharge complex, caused by ponding of water at the surfaces of playa lakes, was investigated at one of the playas currently used for salt harvesting. Here, pore water ion and stable isotope vs depth profiles indicated (1) downward displacement of the original brine body by up to 5 m over 20 years of periodically pumping regional groundwater onto the playa surface, and (2) enhanced downward diffusion of salt through the 30 m thick aquitard, with a time scale of the order of 10^4 yrs. This has implications for long-term management of playa lakes used as evaporation basins, as well as suggesting a possible mechanism for re-distribution of salt in natural groundwater systems that alternate between aridity and higher rainfall conditions.

Declaration

I certify that this thesis does not incorporate without acknowledgement any material previously submitted for a degree or diploma in any university; and that to the best of my knowledge and belief it does not contain any material previously published or written by another person except where due reference is made in the text.

Nikki M. Howes

Publications Associated With This Thesis

Conference Papers

Howes, N.M., Le Gal La Salle, C. and Herczeg, A.L., 2000, **The use of environmental tracers to determine brine leakage from natural groundwater discharge zones and artificial evaporation basins.**, *Proceedings, 4th Environmental Engineering Research Event, Victor Harbor, South Australia, November 21-24, 2000.*

Howes, N.M., Le Gal La Salle, C. and Herczeg, A.L., 2001, **Evidence for brine circulation in a groundwater discharge zone.**, *Proceedings, Water-Rock Interaction 2001, Sardinia, Italy, June 10-15, 2001*, Swets and Zeitlinger, Lisse, Netherlands.

Howes, N.M., Le Gal La Salle, C. and Herczeg, A.L., 2001, **Evidence for brine leakage from a groundwater discharge zone, Raak Plains, Victoria.**, 8th Murray Darling Basin Groundwater Workshop, Victor Harbor, SA, September 2001.

Acknowledgements

The research described in this thesis was carried out collaboratively through the Centre for Groundwater Studies, between the Flinders University of South Australia and CSIRO Land and Water, Adelaide. I would like to thank my supervisors Dr Andrew Herczeg (CSIRO) and Dr Corinne Le Gal La Salle (Flinders) for their continual support, patience, inspiration and technical advice. Thanks also to Andrew and Corinne for the many hours spent reading the chapters of this thesis and the invaluable suggestions for their improvement. I would also like to thank Dr Craig Simmons (Flinders) for his advice and discussions on the hydraulic aspects of the project, as well as for his words of encouragement along the way.

Thanks to John Dighton (CSIRO) for his hard work, support and technical advice in both the field and laboratory, to Leslie McCluskey, Glenn Harrington, Andrew Holub and Martin Wood for their assistance in the field and Megan Lefournour (CSIRO) for her advice to and patience with the numerous students in the laboratory. Thanks to Glenn Harrington (DWLBC) for proofreading chapters and providing useful feedback, and to Andrew Love (DWLBC) for his understanding as I combined finishing a thesis with a new job.

I gratefully acknowledge the financial support of an Australian Postgraduate Award, provided by the Flinders University of South Australia and a supplementary stipend from the Centre for Groundwater Studies. Thanks also to the Centre for Groundwater Studies for assistance with travel costs to attend the 10th International Symposium on Water-Rock Interaction in Villasimius, Italy.

Finally, thanks to my parents Sue and Fred, sister Sally and all those close to me for their encouragement and constant support throughout my studies.

Chapter 1 Introduction

1.1 Scope and Objectives of Thesis

Salinization of surface and groundwater is an increasing problem in arid and semi-arid parts of the world. An increasing population, and the associated expansion of agriculture and industry into arid and semi-arid areas, has placed huge demands on water resources. Schemes that divert water to regions previously adapted to very dry conditions are becoming more and more common, and such interference in these delicate hydrological balances has, in many cases, had disastrous consequences (e.g. Hillel, 1991; Williams, 2001; Vengosh, 2002). For example, rising water tables caused by intensive irrigation and clearing of native vegetation can mobilize salt previously stored in unsaturated zones over many thousands of years, leading to surface and groundwater salinization, and land and infrastructure degradation (Allison et al., 1990).

Salt lakes and playa lakes (a type of dry or episodically filled salt lake) in arid and semi-arid regions can be points of regional evaporative discharge of groundwater and/or surface water. By evapo-concentration of their inflow waters, they also accumulate large quantities of salt in the form of surface and subsurface brines and evaporite mineral deposits. The hypersaline brines can then sink into the underlying sediments, to be stored there or released into the underlying groundwater system, with the potential to further salinize the aquifer (Teller et al., 1982; Duffy and Al-Hassan, 1988; Fan et al., 1997; Wooding et al., 1997 (a&b)). The predominantly groundwater-fed playa lakes (groundwater discharge zones) (Fig. 1.1) are surface expressions of highly saline groundwater systems. Here, water tables are very close

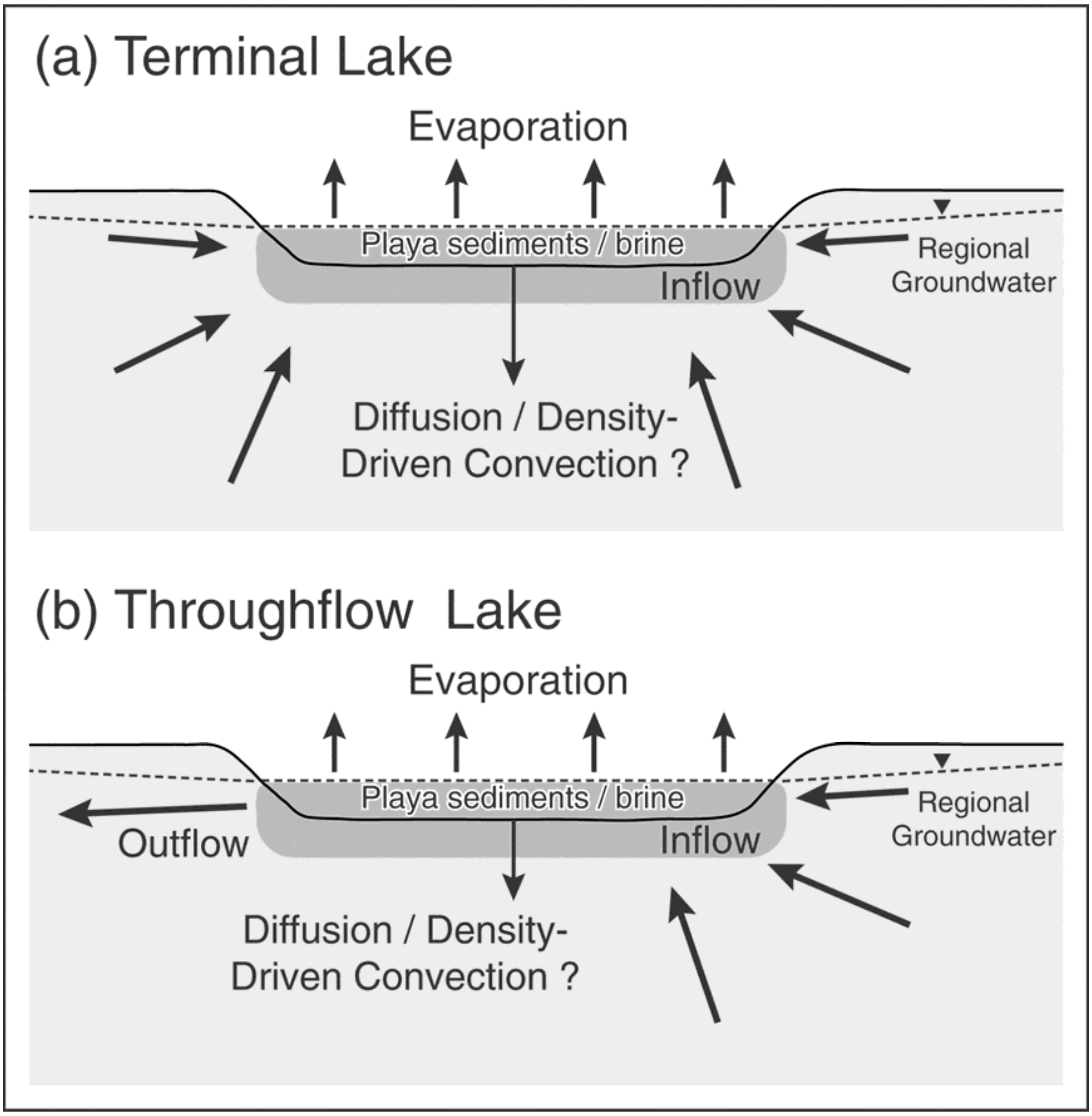

Figure 1.1. Conceptual models of the interactions between groundwater-fed playa lakes (groundwater discharge zones) and the regional groundwater system that feeds them for (a) a terminal lake and (b) a throughflow lake. In both cases, inflowing groundwater is evaporated from a shallow water table under arid and semi-arid conditions. The remaining solutes form a hypersaline subsurface brine that can either remain in the sediments below the playa or leak back into the regional groundwater system via mechanisms such as diffusion, advection (in the case of throughflow lakes) or density-driven convection.

to the surface, typically within about 500 mm, due to either geological constraints or topographic low-points in the landscape. High potential evaporation rates cause groundwater to be evaporated through the thin unsaturated zone, forming the highly saline brines beneath the surface (Fig. 1.1) (Jones et al., 1969). The high salinities of these brines (up to 350 g/L) prohibit the growth of even the most halophytic vegetation, resulting in a bare depression, or “playa”.

Current and fossil salt and playa lakes have been studied for decades because of the roles they play in regional salt and water balances of semi-arid and arid areas, as well as the palaeohydrologic information recorded in their evaporite deposits and shoreline features (e.g. Bowler, 1986; Dutkiewicz et al., 2000; Yechiechi and Wood, 2002). They are also important sources of commercially valuable evaporite minerals, leading to extensive interest in the geochemistry and evolution of their brines. The wide ranges in brine salinity and chemistry observed in salt lake environments are strongly related to the hydraulic processes occurring there and, hence, an understanding of these processes is required to understand their hydrochemistry and evaporite mineralogy (Macumber, 1992).

Much of the current understanding of salt lakes is based on studies of large (>100 km^2), single-lake systems, including examples such as Lake Frome in the southeast of South Australia (Bowler, 1986) and Mono Lake in the USA (Rogers and Dreiss, 1995 (a&b); Phillips et al., 1995). However, complex systems of small (<100 km^2), groundwater-fed playa lakes (groundwater discharge complexes) are also common throughout the world. The degree to which knowledge gained from studies of large salt lakes is applicable to these systems is unclear (Fig. 1.2). In groundwater discharge complexes, the sizes of individual playas, their relative locations, and the presence of recharge (i.e. non-discharge) zones between them may govern their interactions with the underlying groundwater system, as well as influencing the geochemical evolution of their brines. For example, if their chemical compositions are sufficiently different, the relative contributions of regional and locally recharged groundwater sources may determine the chemical evolution of the brines. The magnitudes of these contributions at a particular location in a playa lake may be influenced by the size of the playa, i.e. the proximity of the point to a recharge zone (Fig. 1.3). The location of a playa lake may be important in large groundwater discharge complexes due to variations in the depth to the water table and the resulting differences in the degree of evapo-concentration undergone by the groundwater brine (Fig. 1.3). The presence or absence of an aquitard between the playa lakes and the regional aquifer is also known to be important, controlling brine leakage and inflow from regional groundwater (Chambers et al., 1995) (Fig. 1.3).

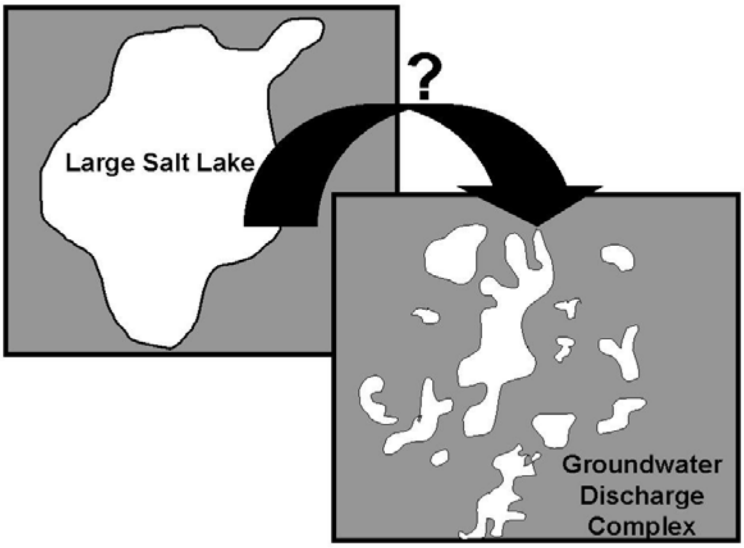

Figure 1.2. Can our understanding of large salt lakes be applied to systems of many small groundwater-fed playa lakes?

In addition to focusing on single large lakes, the majority of studies of salt lake brine hydrochemistry have focused on salt lakes with well-defined surface-water inputs, where the input composition is quite different from that of the resulting brine. Only a few studies, for example of Lake Tyrrell in the central Murray Basin of south-

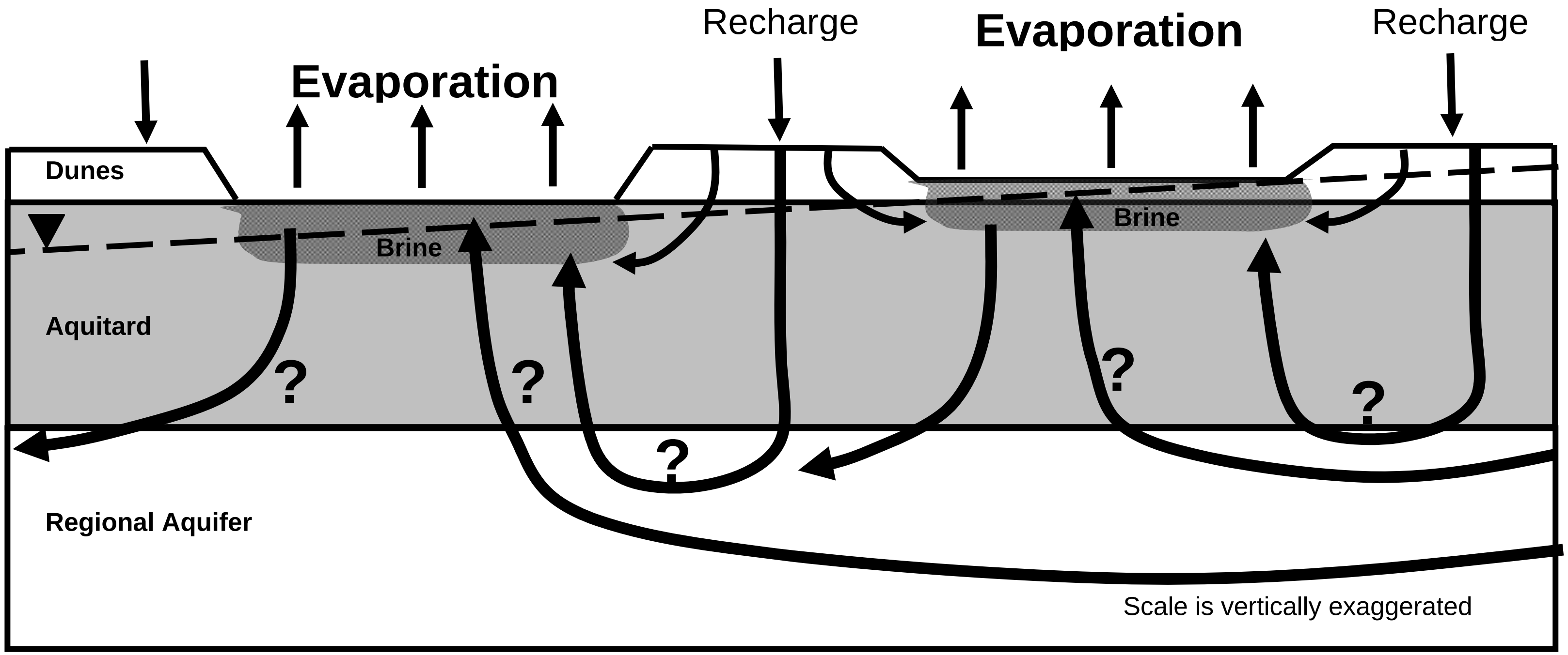

Figure 1.3. Conceptual model of potential groundwater flow patterns below a system of multiple playa lakes separated by areas of net recharge, and underlain by a clay aquitard. The potentiometric surface shown (dashed line) is that of the regional aquifer. Net evaporation of groundwater from the shallow water table forms a hypersaline subsurface brine body below the playa lakes. However, this evaporation rate may vary between playas in large groundwater discharge complexes due to a gradient in the regional potentiometric surface and resulting variations in the depth to groundwater. The water that evaporates may be regional groundwater, locally recharged groundwater, or a mixture of both sources, with the contribution from regional groundwater influenced by the properties of the aquitard (if present) separating the playa lakes from the regional aquifer. The properties of such an aquitard will also control leakage of brine back into the regional aquifer. In large playa lakes, the influence of locally recharged groundwater may vary from the edge to the centre of the playa.

eastern Australia (Herczeg and Lyons, 1991; Macumber, 1992; Herczeg et al., 1992; Hines et al., 1992), the Amadeus Basin in central Australia (Jacobson and Janowski, 1989), Lake Frome in the southeast of South Australia (Draper and Jensen, 1976) and Mono Lake in the USA (Connell and Dreiss, 1995), exist in the literature of brine evolution in predominantly groundwater-fed lakes. These, once again, have focused on single large playas or, at most, the interaction between two playas in a chain of lakes.

Naturally occurring salt lakes can be indicators of the potential long-term effects of salinized land and artificial evaporation basins (Herczeg et al., 1992). In addition to this, they themselves are delicately balanced systems and the imposition of operations, such as salt disposal and salt harvesting, that cause even small changes to their hydrologic balances, may de-stabilize the brine bodies and increase brine leakage (Barnes et al., 1990). These practices, which involve ponding of groundwater at the surface, a step increase in lake level, are becoming increasingly common. In some cases, a substantial aquitard is present, providing the perception that the hypersaline brines are prevented from re-entering the regional aquifer system. However, little observational data are available to confirm this and, in particular, the presence of preferred flow-paths in these aquitards may allow brine migration over greater distances than expected. The long-term impacts of human intervention are difficult to predict when the interactions between playa lakes in their natural states and regional groundwater systems are still not well understood.

The objectives of this thesis are therefore to:

1) Determine the factors influencing the late-stage chemical evolution of the subsurface brines and the resulting evaporite mineral suites in a groundwater

discharge complex. These factors may include (i) the chemical composition (and hence source) of inflow (Hardie and Eugster, 1970), (ii) the properties of the underlying sediments (e.g. the presence / absence of an aquitard) (Chambers et al., 1995; Wood and Sanford, 1990), (iii) playa lake size, (iv) depth to the water table and (v) mineral-solution reactions (Eugster and Hardie, 1978; Eugster and Jones, 1979).

2) Develop a conceptual model for groundwater flow and solute transport below a natural groundwater discharge complex, where a large number of small playa lakes are separated from each other by areas of net recharge and from the regional aquifer by a thick aquitard;

3) Develop a methodology for delineating groundwater flow patterns and identifying brine leakage below groundwater discharge complexes.

4) Determine the impact of increased lake levels (e.g. due to salt disposal or salt harvesting) on brine leakage below natural playa lakes underlain by a clay aquitard.

1.2 Methodology

The Raak Plain groundwater discharge complex, located in the central Murray Basin of south-eastern Australia, was selected as the study site for this investigation, due to the fact that it contains a large number (> 50) of small (< 100 km^2) playa lakes. Being located at the centre of a large regional groundwater basin, it is also believed to discharge saline (Total Dissolved Solids > 30 g/L) regional groundwater through a 30 m thick clay aquitard. This study adopts a combined approach, using present hydraulic data (water levels and aquifer properties) that give a snapshot of the hydraulic processes currently operating below the discharge complex, and environmental tracers that integrate processes over long time scales. Data collection and interpretation at both the individual playa and discharge complex scales allows assessment of the spatial variability in processes at both these scales. A suite of chemical and isotopic tracers, including major ion chemistry, $^{2}H/^{1}H$ and $^{18}O/^{16}O$, $^{13}C/^{12}C$ and ^{14}C, and ^{36}Cl, is utilized to better constrain the large range of hydrochemical and physical processes potentially occurring in the groundwater discharge complex. One particular issue in evaporitic environments is de-coupling between the solute and water systems during the evaporation process, which removes water and leaves solutes behind. In this situation, ionic species can be used as tracers of the solutes, whereas the stable isotopes of water, $^{18}O/^{16}O$ and $^{2}H/^{1}H$, are ideal tracers of the water molecule. The radioactive tracers, ^{36}Cl and ^{14}C are used to determine the time scales of solute transport below the discharge complex.

1.3 Site Description

1.3.1 The Murray Basin, South-Eastern Australia

The hydrological balance of the Murray Basin of south-eastern Australia (Fig. 1.4) has been modulated over the last several hundred thousand years by alternating periods of wet (high groundwater levels) and semi-arid to arid (low groundwater levels) climatic conditions (Bowler and Wasson, 1984). At present, the climate in the Basin is semi-arid (rainfall ~ 300 mm/y and potential evapotranspiration ~ 2000 mm/y), and the shallow aquifer system in many parts is saline. The most saline groundwaters occur at the centre of the Basin, where they are discharged either via the River Murray or by evaporation from shallow water tables in groundwater discharge complexes (Brown, 1989; Evans and Kellett, 1989). In the latter case, the residual salts form hypersaline groundwater brines (> 250 g/L). Today, there are many active and “fossil” (currently inactive) groundwater discharge complexes, with associated brine bodies, scattered throughout the centre of the Murray Basin.

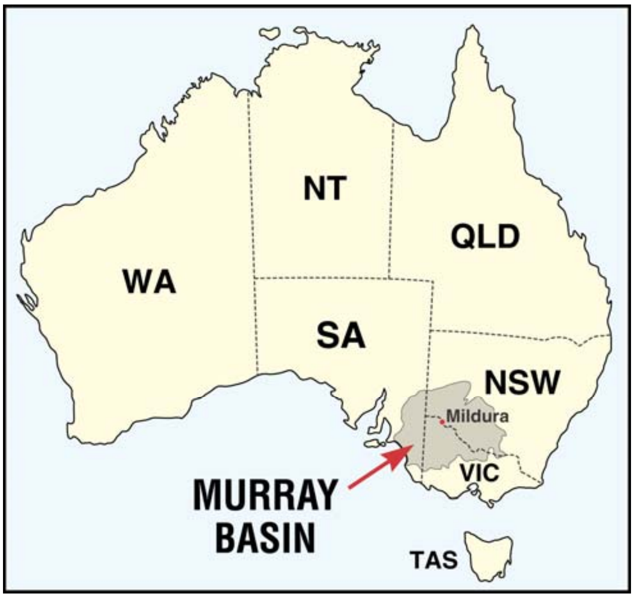

Figure 1.4. Locality map of the Murray Basin in south-eastern Australia.

Rising water tables, caused by clearance of native vegetation, are now bringing saline groundwater to the surface, degrading agricultural land and increasing the rate of discharge of saline groundwater to the River Murray. To alleviate the problem, a number of groundwater interception schemes currently divert saline groundwater to natural playa lakes or man-made basins. These are used as evaporation basins for the storage of saline brines, or for removal of water for salt harvesting. Many of the natural playa lakes are underlain by clay aquitards, making them attractive sites for such operations.

1.3.2 The Raak Plain Groundwater Discharge Complex

The Raak Plain groundwater discharge complex is located in the central Murray Basin, in the Mallee region of western Victoria (Figs. 1.5 and 1.6). It is the largest

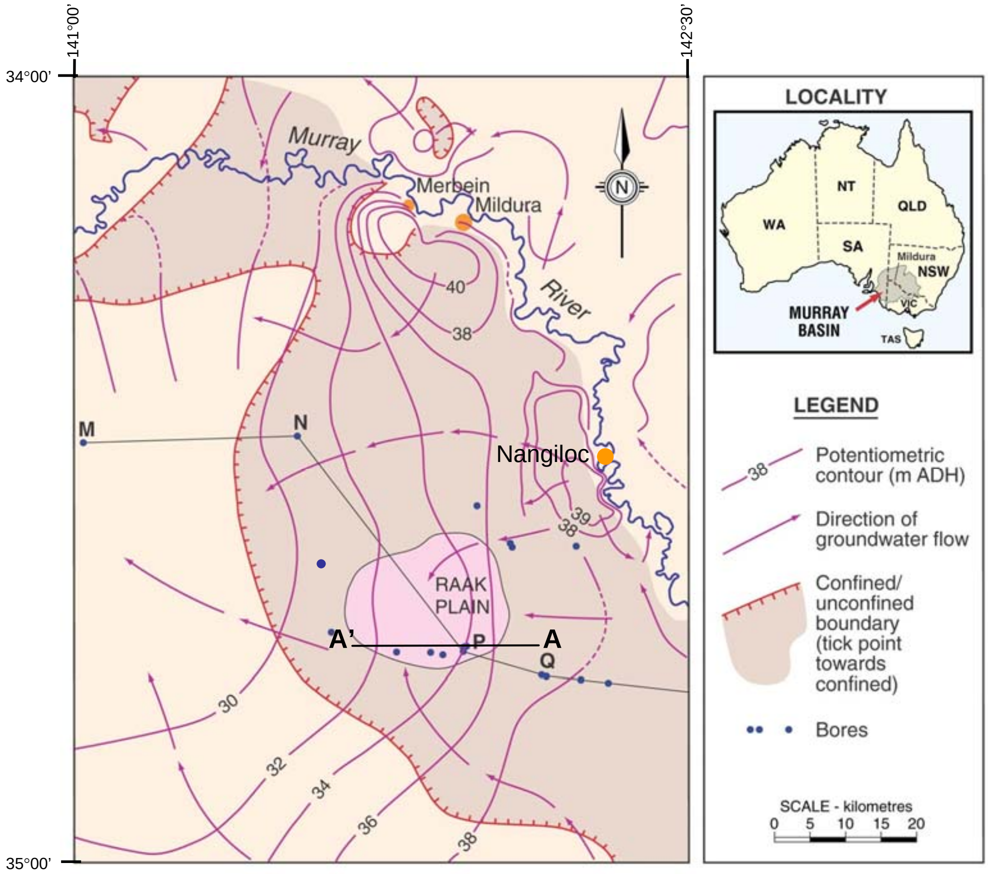

Figure 1.5. Hydrogeological map of Raak Plain and surrounds, showing the extent of the confining Blanchetown Clay aquitard, potentiometric contours for and locations of bores in the Parilla Sand aquifer (after Rural Water Commission, 1991). Cross section MNPQ is discussed in Section 1.2.3 in terms of the regional geology and hydrogeology. Transect AA' is the cross-section along which this study is focused.

groundwater discharge complex in the Murray Basin, covering an area of approximately 400 km^2, and containing more than 50 small playa lakes (Fig. 1.6). Varying physical characteristics of the playa lakes (size, shape, presence/absence of surface water) were expected to cause differences in brine evolution, and in turn evaporite mineralogy, supported by the fact that some playas contain a surface salt crust whilst others do not. The playa lakes are also separated from the regional aquifer by the 30 m thick Blanchetown Clay aquitard. It was anticipated that this would greatly affect groundwater and solute movement below the discharge complex, and allow extension of previous work on playa lakes and man-made

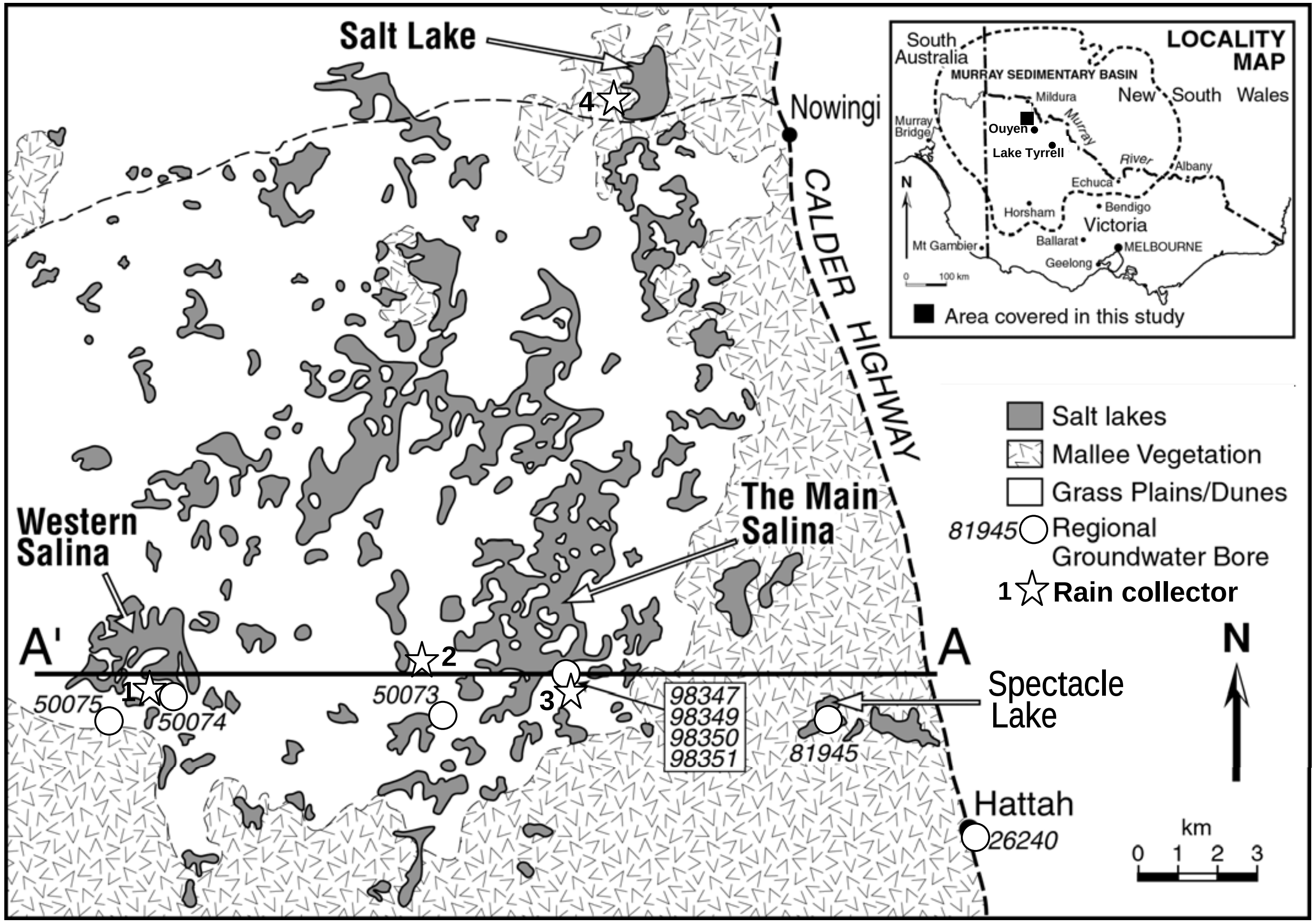

Figure 1.6. Site map of the Raak Plain groundwater discharge complex, showing locations of the playa lakes that are the focus of this study, the bores screened in the Parilla Sand aquifer and transect AA' along which the study is also focused. The regional groundwater flow direction is approximately from east to west. Bulk rainfall / dry deposition collector sites are also shown as stars with site numbers (i.e. 1 = Rain1).

evaporation basins that are in direct connection with the regional aquifer (e.g. Jacobson and Jankowski, 1989; Macumber, 1992; Herczeg et al., 1992; Connell and Dreiss, 1995; Simmons and Narayan, 1998).

The Victorian Mallee region, in which Raak Plain is located, is characterised by a predominantly flat landscape, overlain by systems of transverse dunes. These dunes, which separate the playa lakes at Raak Plain, are usually densely vegetated with mallee, a collective term for the shrubby trees and low-lying scrub or heath that exists mainly in low rainfall areas (250 – 450 mm/y) (Walker et al., 1992). Mallee trees have slender trunks stemming from a single lignotuber just below the surface and are well known for their efficient use of water leading to low recharge rates (Cook et al., 1996; Walker et al., 1992). The landscape of the groundwater discharge complex, consisting of playas intermingled with sand plains, samphire-vegetated gypsum flats and bordering dunes, was described by Macumber (1983, 1991) as a single landform, the "boinka".

The boundary of the Raak Boinka, with its sparsely vegetated floor approximately 15 to 20 m below the surrounding Mallee dunefields, can be easily identified from aerial photos. The lake-bed elevations are approximately 32 to 34 m AHD, with the surrounding plains and dunes ranging between 35 and 55 m AHD (Sheet 7328 (Edition 1), 1977, National Topographic Map Series). A network of interconnected salinas forms the Main Raak salina in the eastern half of Raak Plain, as described by Macumber (1991) and referred to as the Main Salina in this study (Fig. 1.6). One of the smaller salinas (Spectacle Lake), at the eastern boundary of the boinka, supports a commercial salt harvesting operation. The salinas are active groundwater discharge zones, with brines just beneath the surface reaching chloride concentrations of up to 200 g/L (Howes, 1998). There are no surface water inflows to the playa lakes, although the water table is near or above the playa surfaces. The only input of water is from groundwater in-flow, direct rainfall and some minor runoff from their immediate surroundings (Macumber, 1991). A slight east-west gradient in water table elevation reflects the regional groundwater hydraulic gradient of approximately $1{:}10^4$ in the underlying Parilla Sand aquifer.

Land use in the eastern three quarters of Raak Plain consists mainly of dryland agriculture and light grazing, with the western quarter forming part of the Murray Sunset National Park. Although anecdotal evidence suggests that Raak Plain was never cleared of native vegetation, there is evidence of rising water tables from land clearing and increased irrigation in surrounding areas. Salinized land, identified by changing vegetation from grasses to salt-tolerant samphire vegetation, can be seen encroaching on roads and fencelines.

The climate in the central Murray Basin is semi-arid (Macumber, 1991), with potential evaporation exceeding rainfall by roughly an order of magnitude (the mean annual precipitation is 250 - 350 mm/y compared with a mean annual potential evaporation of around 2000 mm/y (Allison et al., 1988)). Long-term average rainfall data for stations at Ouyen (approximately 30 km south of Raak Plain) and Mildura (approximately 60 km north) is available from the Bureau of Meteorology website, (Bureau of Meteorology, Australia, 2002). Rainfall in the Mallee is scattered throughout the year, but is mostly concentrated in the period between May and September (winter). Graphs of average monthly rainfall and number of raindays are included as Appendix A (Bureau of Meteorology, Australia, 2002). The mean annual rainfall is 290 mm at Mildura and 335 mm at Ouyen. Raak Plain lies between these two stations and the mean at this location is therefore expected to be approximately 300 mm/y. Rainfall is fairly evenly distributed throughout the year, with most monthly averages being greater than 20 mm (Appendix A). The driest months are March and April and the wettest month is October.

1.3.3 Geology and Hydrogeology

The 3 x 10^5 km^2 Murray Basin, part of the larger Murray-Darling Basin, is a low-lying, saucer-shaped basin with a surface drainage system consisting solely of the River Murray and its tributaries. Groundwater flows from up-basin recharge areas to zones of groundwater discharge (playa lakes) in the semi-arid centre, and ultimately to the River Murray. The basin consists of up to 600 m of unconsolidated Cainozoic sedimentary rocks that are the result of successive marine transgressions in the Oligocene – Early Miocene and Late Miocene – Early Pliocene times. These form at least four major regional aquifer systems – the Renmark Group, Murray Group and Pliocene Sands aquifer systems and the Shepparton Formation partial aquifer system (Brown and Radke, 1989; Lawrence, 1975) (Fig. 1.7). In the western half of the Murray Basin, in the vicinity of Raak Plain, the Renmark Group is overlain by the Murray Group, which includes the Duddo or Murray Group Limestone. This is up to 130 m thick and is overlain by the Bookpurnong Beds, a clay layer up to 100 m thick deposited during a marine transgression (Macumber, 1991, 1992).

The Parilla Sands Aquifer (also known as the Loxton or Pliocene Sands Aquifer in South Australia) is the main unconfined to semi-confined aquifer over the western part of the Murray Basin. It is a 60-70 m thick layer of unconsolidated to partially consolidated sands and is semi-confined when overlain by thicker deposits of the Plio-Pleistocene Blanchetown Clay, a sequence of gypsiferous clays, silts and minor fine sands (Macumber, 1992; Allison et al., 1985; Rogers, 1995). Aeolian reworking of the Parilla Sands and Blanchetown Clay has caused the linear dunefields (Woorinen Formation) that cover much of the Mallee (Macumber, 1992). At the top of the profile are the playa deposits (Yamba Formation), consisting of lacustrine gypsiferous clay and gypsum-quartz sand mixtures, associated with gypsiferous dunes, in the bed of the former Lake Bungunnia, the inland lake that once covered the central mallee region (Rogers, 1995).

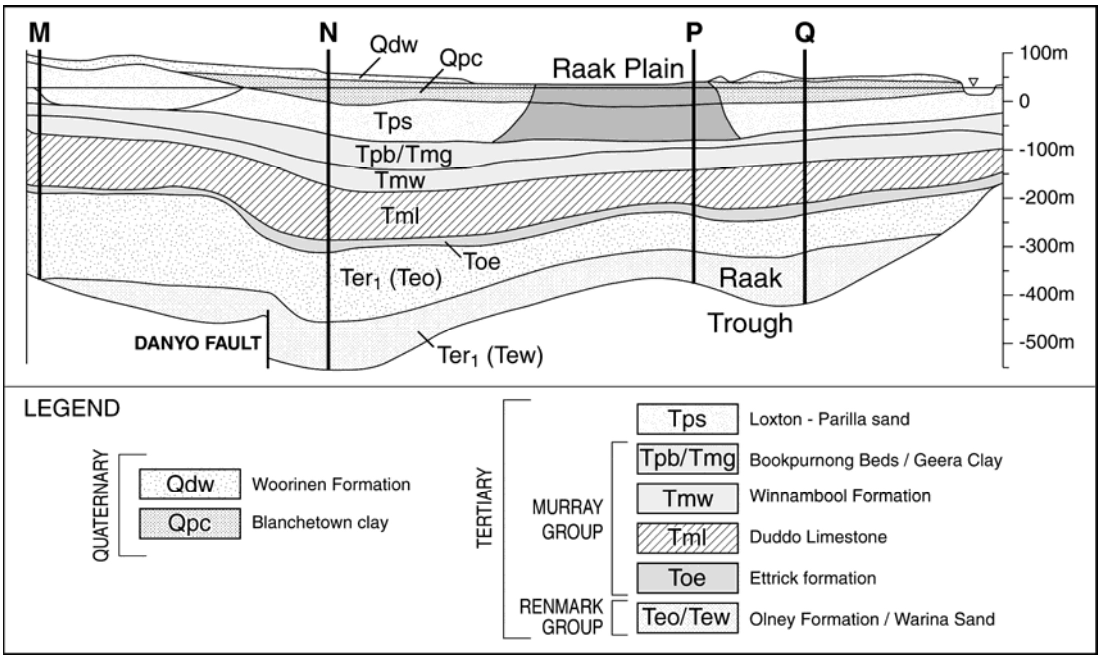

Figure 1.7. Cross section showing the three main aquifer systems in the vicinity of Raak Plain, the Renmark Group, Murray Group and Parilla Sand aquifer systems (after Rural Water Commission, 1991). The location of transect MNPQ is shown on Figure 1.5.

The groundwaters of the central Murray Basin contain dissolved solutes that are of predominantly oceanic origin. Marine aerosols deposited at the land surface on a basin-wide scale are recharged predominantly by winter rains (Jones et al., 1994; Herczeg et al., 2001). Hence, groundwater compositions are remarkably similar to that of seawater, with only slightly higher calcium, bicarbonate, sulphate and silica concentrations, and slightly lower potassium due to mineral-solution reactions during its evolution (Macumber, 1991). The hydrochemically well-evolved nature of these groundwaters (Herczeg et al., 2001) provides the opportunity to study the late-stage hydrochemical evolution of brines as they become evapo-concentrated in playa lakes.

Groundwater flow in the Parilla Sand, as well as the underlying units, in the vicinity of Raak Plain is from east to west. However, as the discharge complex is a groundwater sink, the local flow system in the Parilla Sands aquifer may be directed radially inward towards it (see Fig. 1.5). Very little potentiometric data is available in this region to confirm this. Values for hydraulic conductivity (2 m/day), hydraulic gradient (0.2 m/km to 0.3 m/km) and groundwater flow velocity (0.001 m/day to 0.005 m/day) in the Parillla Sand aquifer below the Mallee region are given by Macumber (1991). The groundwater discharge status of Raak Plain can be seen in the standing water levels of the bores screened in the Parilla Sand aquifer, which are up to 2 m above the playa floors. Hence, the Parilla Sand aquifer is believed to be a major source of salt and water to the playa lakes of Raak Plain.

1.3.4 Previous Investigations at Raak Plain

Macumber (1991, 1992) conducted a study of surface water/groundwater interactions in northern Victoria, and included Raak Plain as a study site. He observed that the salinas are occasionally covered by a shallow sheet of water in winter, but dry out in the warmer months, and follow a seasonal cycle of halite dissolution and re-precipitation. He also proposed that each salina may be categorized as either halite producers, with a high Cl/Br ratios when wet, or as through-flushing systems, with lower Cl/Br ratios. By comparing the hydrochemistries of some of the lakes of the Raak boinka he also suggested that the lake waters are simply concentrated versions of the regional groundwater, and that the groundwater brines and their parent lake waters have compositions pre-determined by the composition of this source. He noted that, although the shallow groundwaters beneath the salina floor are close to halite saturation, very few salinas are halite producers, indicating that just the presence of the shallow brine is not sufficient to produce a halite evaporite body at the surface.

1.3.5 Characteristics of the Playa Lakes Selected for This Study

Locations of the playas included in this study are represented in Table 1.1 by their distances from the eastern margin of the discharge complex (designated here as the Calder Highway), approximately along the regional groundwater flow path (transect AA'; Fig. 1.6). They cover a range of sizes (10^4 m^2 to 10^7 m^2) and locations, and include some that contain a surface water body and salt crust, as well as dry salt pans (Table 1.1). Spectacle Lake was chosen because it now acts as a series of evaporation ponds for a salt harvesting operation, and is an example where the hydraulic balance of the lake has been altered by the ponding of groundwater pumped onto the playa surface from the Parilla Sand aquifer.

The shallow (< 3 m deep) sedimentary sequences in the lakes at Raak Plain generally comprise a 20 to 50 cm layer of chocolate-brown silty fine sand (the Yamba Formation playa sediments) (Appendix B). This may be covered by a surface salt crust or thin algal mat, depending on the conditions in the lake. Algae can grow on the playa floors in winter, when long periods of rain maintain temporary pools of comparatively fresh water in the lower-lying parts of the lakes. The compositions of the playa sediments vary slightly between playas, but are generally dominated by fine quartz sand, with some kaolinite, muscovite and potassium- and sodium-feldspar (see Appendix B).

Table 1.1 Summary of general lake characteristics. Water table information is from piezometers installed in playa sediments. Maximum chloride concentration of the brine is from pore water extracted from shallow playa sediments.

	WESTERN SALINA		MAIN SALINA		SALT LAKE	SPECTACLE LAKE (HARVESTED)	
Approximate Area (km^2)	4		15		1.1	0.4	
Shape	Irregular		Irregular		Sub-rounded	Sub-rounded	
Distance From Eastern Margin (km)	17		8.5		2.5 (located in the north of the discharge zone)	2.2	
Surface Water Catchment	Lake surface. Some sand dunes on western margin. Poorly vegetated.		Lake surface. Some sand dunes scattered around margin. Poorly vegetated.		50-100 m beyond lake margin. Well-vegetated (mallee & sheoak) sand dunes on all sides.	50-100 m beyond lake margin. Well-vegetated (mallee) sand dunes on all sides.	
Salt Crust?	No		No		Yes – dissolves in winter	Yes, controlled.	
Surface Water?	Rare - only after heavy rain		Rare - only after heavy rain		Yes - up to 15 cm deep	Controlled application. Pumped from Parilla Sand aquifer.	
Site Name	West1	West2	Main1	Main2	Salt1	SM5	SM1
Approximate Distance From Shore (m)	100	20	100	20	50	50	5
Sample Depth (m below ground)	1.2	1.25	1.95	0.5	0.5		1.7
Lake Floor Elevation (m AHD)	32.1	32.2	32.5	32.5	na	na	na
Hydraulic Head (m AHD)	31.6 to 32.3	31.9 to 32.5	32.6 to 32.9	32.2 to 32.5	na	na	na
Hydraulic Head (m above ground)	-0.5 to 0.2	-0.3 to 0.3	0.1 to 0.4	-0.3 to 0.0	0.5	na	0.95 to 1.1
Max. Pore Water salinity (mg/kg TDS)	242 500	234 000	243 800	260 700	259 200	250 200	251 800

The playa sediments are underlain by a tight grey silty, clayey fine sand, consisting mainly of quartz with kaolinite and Fe-oxyhydroxides (Appendix B). The clays fill the space between the quartz grains and the Fe-oxyhydroxides form coatings on the grains, in some cases forming a weak cement (Appendix B). Iron oxyhydroxides occur heterogeneously in the sediment profiles, ranging from small (<0.5 cm) red and yellow zones and pebbles, up to large chunks of ironstone and entire depth intervals of red or yellow colour. The increased clay content and dramatic colour change from brown to grey, red and yellow indicates the transition from playa sediments to the top of the Blanchetown Clay (Fig. 1.8). Mottling occurs in well-defined zones corresponding to past water table levels. Variations between playas in evaporite mineralogy and the degree of iron mineralization are described in Chapter 3.

Figure 1.8. Trench cut into playa surface at Western Salina, showing ferruginization in the Blanchetown Clay. The brown sediments in the top 20 cm are the playa sediments (Yamba Formation).

1.3.5.1 Western Salina

Western Salina is located near the western margin of Raak Plain (Figure 1.6), and is a large ($\cong$ 4 km^2), irregularly shaped salina (Table 1.1). The playa is normally dry, although a small ponded surface water body, less than 10 cm deep, occasionally forms after heavy rainfall (see Appendic C). This water blows across the surface of the playa with the prevailing wind and is eventually re-evaporated or infiltrates to the shallow water table.

Western Salina is surrounded by flat grassy plains, approximately 2 m above the playa surface, except along the western boundary of the playa, where sand dunes, up to 20 m high and sparsely vegetated with mallee, are present (see Appendix C). Recharge through these dunes is expected to contribute, along with direct rainfall onto the playa surface, to the salt and water balance of the lake, particularly during high rainfall events. The lack of a significant surface water body in this playa, however, is reflected in its irregular shape, as discussed in Section 1.3.5.5. The hydraulic head in a piezometer screened 1.2 m below the playa surface fluctuates between 50 cm below and 20 cm above ground. There is no surface salt crust, however a temporary efflorescence of tiny white crystals can often be observed on the playa floor after rain, usually within 20 m to 50 m of the playa edge.

1.3.5.2 The Main Salina

The Main Salina is the large salina stretching north-south along the eastern side of Raak Plain, with an area of approximately 15 km^2 (Fig. 1.6; Table 1.1). Surface characteristics are very similar to those of Western Salina (Appendix C). Surface water occurs periodically after heavy rain, only as a thin film that blows across the lake surface and eventually evaporates or infiltrates to the shallow water table (Appendix C). The surroundings of the lake are similar to those of Western Salina, with extensive grassy plains and low sand dunes sparsely vegetated with mallee and sheoak trees (Appendix C). The piezometric surface, reflected by the playa surface elevation, is approximately 40 cm higher than at Western Salina, which is expected due to the westerly trending hydraulic gradient across the entire discharge complex. Hydraulic heads in a piezometer at the centre of the lake fluctuate seasonally between 0.1 and 0.4 m above the playa surface (Table 1.1). Surface efflorescences were occasionally observed on the playa surface at the Main Salina after periods of rain, despite no substantial salt crust. These are normally most prevalent within 20 to 50 m of the edges of the playa, as at Western Salina.

1.3.5.3 Salt Lake

Salt Lake is the only playa lake included in this study that does not lie along the east-west transect AA’ (Fig. 1.6). It is the smallest of the three natural lakes selected for this study, with an area of 1.1 km^2, and lies on the north-eastern border of Raak Plain, where surrounding land is now being cropped for dryland agriculture (Fig. 1.6, Table 1.1). It is 2.5 km from the eastern boundary of the discharge complex and is buffered from the surrounding cropped areas by a 50 m wide strip of mallee and sheoak trees (Table 1.1, Appendix C). The natural physical setting of Salt Lake would have been different from those in the interior of Raak Plain prior to land clearance. It is at the margin of the discharge complex, where the landscape is undulating, with east-west trending sand dunes that are densely vegetated with mallee trees (see Section 1.3.2). This is in contrast to the centre of the discharge complex, which is comparatively flat and bare of vegetation, due to the high salinity of and shallow depth to groundwater (see Section 1.3.2).

The lake edge at Salt Lake has a steeper slope than at Western Salina and the Main Salina and surface water, either from groundwater discharge or direct rainfall, can be up to 15 cm deep, particularly around the edges of the lake (Table 1.1, Appendix C). This surface water body diminishes in summer due to evaporation, and the centre of the lake becomes dry whilst the remaining water is blown around the lake.

During each trip to the site, there was a surface salt crust up to 5 cm thick covering much of the lake surface (Appendix C), but this almost completely dissolves in the height of winter. Large cracks were occasionally observed in the salt crust, forming large polygons, approximately 2-3 m wide (Appendix C). At the single piezometer site at the centre of the lake, the hydraulic head 2.6 m below the lake surface has been constantly 0.5 m above ground level, and did not fluctuate as much as in the other lakes (Table 1.1). Ground and hydraulic head elevations at Salt Lake could not be determined relative to the AHD, as there was no known reference point in the vicinity.

1.3.5.4 Spectacle Lake

Spectacle Lake is located 2.2 km from the eastern boundary of Raak Plain and is the smallest of the lakes included in this study (0.4 km^2) (Table 1.1, Fig. 1.6). Regional groundwater, pumped from the Parilla Sand aquifer, is currently ponded on the surface of the lake and evaporated to enhance the precipitation of evaporite minerals (Appendix C). Anecdotal evidence indicates that, before human intervention, the lake was similar to Salt Lake in that it always supported a surface water body and salt crust (Duncan Thompson, Hattah Salt, pers. comm., 2001). The natural physical setting of Spectacle Lake is also similar to that of Salt Lake, as it is surrounded by mallee-vegetated sand dunes (Fig. 1.6; Appendix C).

Spectacle Lake has been harvested for its naturally produced evaporite minerals from as early as 1900. The first production bore was installed in 1980 to pump water from the Parilla Sand aquifer onto the surface of the lake to enhance the production of salt by solar evapo-concentration. The base of the lake was not lined to contain the brine due to the presence of more than 30 m of Blanchetown Clay directly underlying the lake. Groundwater is now pumped from bores around the edges of the lake into a series of three evaporation basins. Water is first pumped into Pond 1 for volume reduction in late autumn and spring, and iron oxyhydroxides and $CaCO_3$ are removed from solution as precipitates. This water is then allowed to flow into Pond 2 for further evapo-concentration and gypsum ($CaSO_4 \cdot 2H_2O$) precipitates. When the specific gravity of the water reaches 1.216, the water is pumped into the smaller Pond 3 where halite (NaCl) is crystallized (Appendix C). This halite is contaminated with iron, causing it to be granular in shape and brownish in colour. Finally, the remaining bittern is pumped into the low volume crystallizers, where the high value minerals such as epsomite ($MgSO_4 \cdot 7H_2O$), bischofite ($MgCl_2 \cdot 6H_2O$) and sylvite (KCl) are extracted (Appendix C). The mineral extraction rate at Spectacle Lake was 2 000 to 3 000 tonnes per year between 1982 and 1996, and 4 000 to 6 000 tonnes per year from 1996 to present. The ponded surface water in Ponds 2 and 3 can be up to approximately 50 cm deep, although both are allowed periodically to dry out for harvesting (Appendix C).

1.3.5.5 Surface Water vs Dry Lakes

Bowler (1986) classified Australian lakes in terms of their current hydrologic stage of evolution, with each type distinguished by its surface morphology. The types range from Type A, with a permanent, relatively deep surface water body of low salinity, to Type E basins, with little surface-water influence that are dominated by groundwater processes such as groundwater bevelling and salt-weathering. The types are distinctive in their morphology, particularly their plan outlines, due to different relative influences of surface- and groundwater processes such as wave action and salt weathering respectively. Irregular plan outlines are caused by playa migration into inter-dunal corridors via salt weathering, the destabilization of sediments by surface salt efflorescences that make them prone to aeolian deflation (Bowler, 1986). Lakes that support even a shallow surface water body for much of the year tend to have smoother outlines as a result of wave action. Based simply on their surface physical characteristics and the classification scheme of Bowler (1986), the lakes at Raak Plain can be divided into two categories:

Salt Lake and Spectacle Lake fall into the category of Type D basins, ephemeral and high salinity, with comparatively regular plan outlines (Fig. 1.6). Some other examples of Australian Type D lakes are Lakes Eyre and Tyrrell. According to the description of Bowler (1986), under prolonged dry conditions in basins of this type, the surfaces may dry sufficiently to permit salts to effloresce. Salt Lake (and probably Spectacle Lake in its natural state) has a thin layer of surface water for most of the year, which dries up only in the driest summer conditions, and a surface salt crust that is partially re-dissolved in the winter rains.

Western Salina and Main Salina can be categorised as Type E basins, with little surface-water influence, and hence irregular plan outlines. They rarely contain ponded surface water and never a surface salt crust, with the exception of minor surface efflorescences. These playas expand laterally via salt weathering as described above, and are modified by episodic aeolian deflation, which forms “islands” within the dry lakes (Bowler, 1986). Some other examples of Type E basins in Australia are Lakes Amadeus, Gairdner and Torrens.

1.4 Preliminary Conceptual Models of the Playa Lakes at Raak Plain

A series of preliminary conceptual models can be constructed for the water and solute balances of the playa lakes at Raak Plain, based on a general understanding of the hydrogeology (Fig. 1.9 (a-d)). The geology consists of the regional Parilla Sand aquifer, the confining Blanchetown Clay unit, the thin layer of playa sediments in the lake bed and the dune / plain Woorinen Formation at the margins of the lake. For the purpose of this study, no interaction between the Parilla Sand and the underlying Geera clay is considered. In the conceptual model, the Blanchetown Clay is divided into the Upper and Lower Blanchetown Clay, with the Upper Blanchetown Clay being that depth interval containing the brine body.

The potential sources of water and salt that undergo evaporation in the lakes and ultimately form brines are groundwater discharged from the Parilla Sand aquifer, recharge through the adjacent sand dunes, and direct rainfall onto the lakes. The two scenarios for the fate of the brine body that is formed below the lake are a) confinement within the Blanchetown Clay by the low conductivity of the aquitard and the upward pressure exerted by the Parilla Sand aquifer, or b) transport of solutes by some mechanism back into the Parilla Sand aquifer.

Possible conceptual models can therefore be characterized by two end-members, the Isolated Systems Model (Fig. 1.9a) and the Connected Systems Model (Fig. 1.9(b-d)). In the Isolated Systems Model, there is no interaction between the playa lake and the regional groundwater system. The main sources of solute and water are direct rainfall onto the lake or recharge through the adjacent sand dunes, and the brine body is confined within the Blanchetown Clay. In the Connected Systems Model, there is some exchange of solutes between the regional groundwater system and the playa lake and inputs from direct precipitation or local recharge are negligible. In one possible scenario, the dominant source of solute and water is upward discharge from the Parilla Sand aquifer, but salt is accumulated in the subsurface brine body and does not re-enter the aquifer (Fig. 1.9b). The second possible Connected Systems scenario is similar, but with solutes re-entering the aquifer by either advection (Fig. 1.9c) or diffusion (Fig. 1.9d). Real playa lake systems represent a continuum between these conceptual models.

A water and solute balance can be defined for each of the above conceptual models. For the water balance, the assumption is made that the hydraulic systems of the natural lakes are at steady state, based on the fact that water levels in the lakes are maintained approximately at the lake floor by evaporation and do not fluctuate significantly over large time scales. Under these conditions, the sum of inputs, i.e. direct rainfall onto the playa surface (R_p), inflow from peripheral dunes (I_d) and upward leakage from the Parilla Sand (I_{PS}) equals the sum of outflows, i.e. evaporation (E) and advective leakage to the Parilla Sand (L):

$$R_p + I_d + I_{PS} = E + L \qquad (1.1)$$

For solutes, the brine body may be either at steady state (sum of solute inputs equal to the sum of outputs), in a state of net accumulation (inputs > outputs) or in a state of net loss (inputs < outputs). In the case of the Isolated Systems model (Fig. 1.9a), net accumulation is inferred unless there is loss of efflorescent salts from the lake surfaces by deflation (Bowler and Wasson, 1984; Bowler, 1986; Wood and Sanford, 1995). This is also the case for the first Connected Systems Model. However, for the two Connected Systems Models where solute is lost to the Parilla Sands aquifer

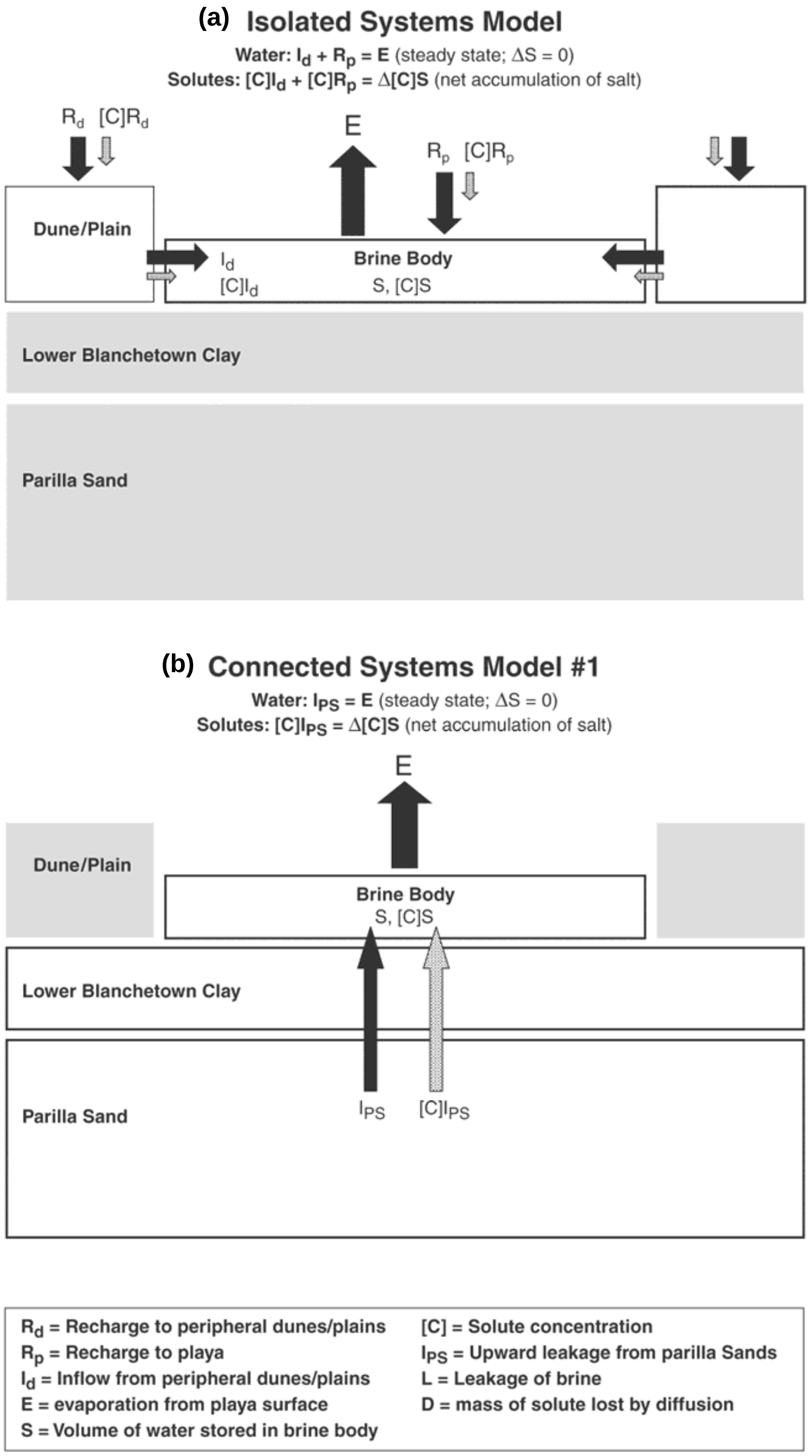

Figure 1.9. Preliminary conceptual models of the water and solute balances of the playa lakes at Raak Plain: (a) An Isolated Systems model, assuming negligible interaction between the playas and the regional Parilla Sand aquifer. In this model, the major sources of water and solutes to the brines are local recharge through adjacent dunes and plains and direct rainfall deposition and there is no brine leakage into the Parilla Sand. (b) A connected systems model assuming that regional Parilla Sand groundwater is the major source of solutes and water to the playas, but that leakage of the brine does not occur.

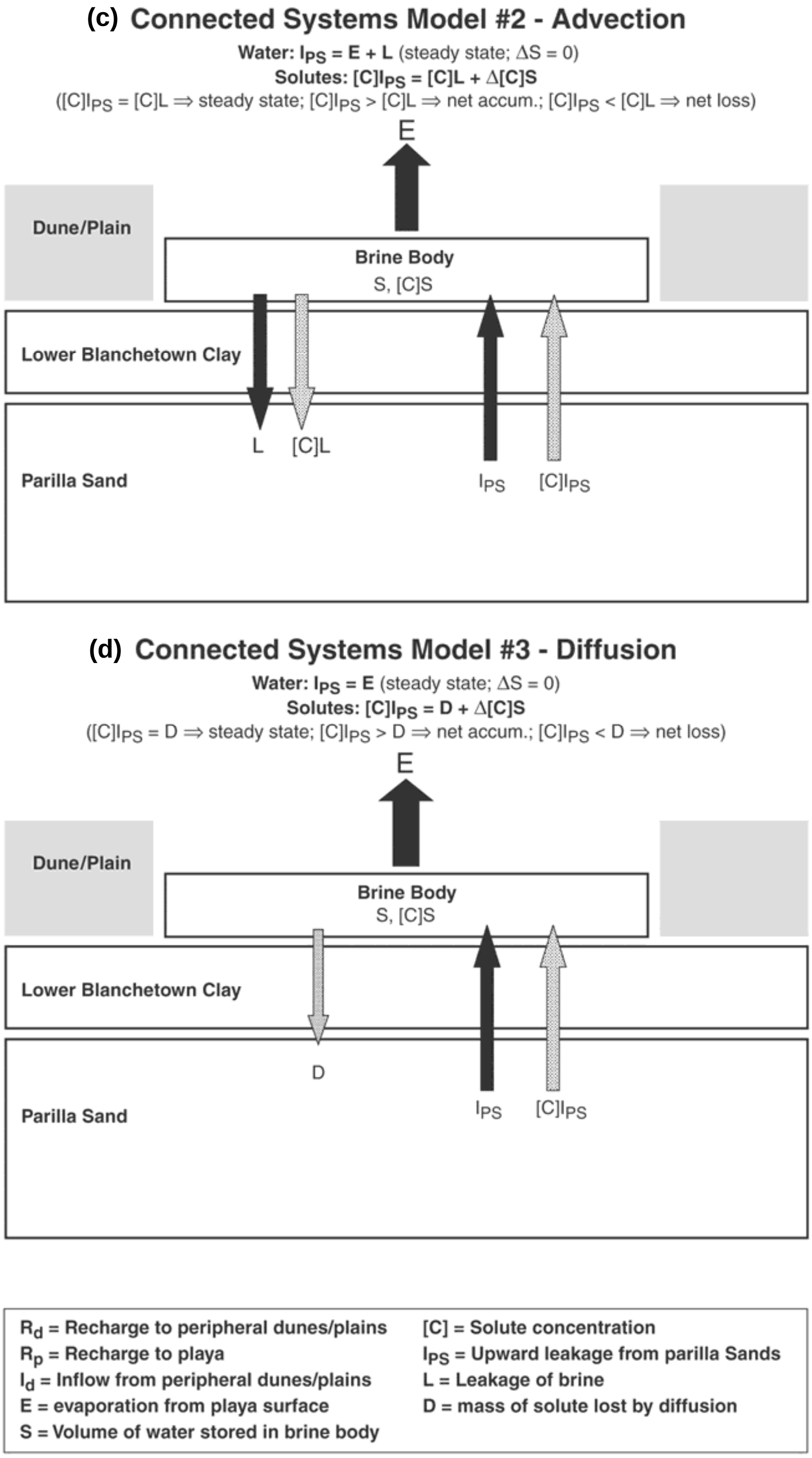

Figure 1.9 (continued). Preliminary conceptual models of the water and solute balances of the playa lakes at Raak Plain: (c) A connected systems model incorporating advective leakage of brine. (d) A connected systems model incorporating leakage of solutes from the brine body by diffusion only.

via either advection or diffusion, the brine body may be at steady state, or experiencing a net loss or accumulation, depending on the magnitudes of the fluxes.

These conceptual models will be used as a basis for interpreting the results of the environmental tracer and hydraulic studies of the playa lakes at Raak Plain, which are described in the following chapters. Chapter 3 investigates the physical and mineralogical controls on the chemical evolution of the subsurface brines at Raak Plain, and what this can tell us about groundwater flow and solute transport below the discharge complex. Hydraulic data are used in Chapter 4 to develop a conceptual model of groundwater flow below the groundwater discharge complex and a numerical model is used in Chapter 5 to test the validity of this model and its sensitivity to various parameters. Chapter 6 compares conclusions drawn from the isotopic tracer signatures of the brines and groundwaters at Raak Plain with the conceptual model derived in Chapter 4 in order to improve confidence in and refine this model. Chapter 7 focuses specifically on processes occurring below Spectacle Lake, and how they relate to the change in the hydraulic balance of the playa by the imposition of a salt harvesting operation.

1.5 Overview of Previous Work on the Hydrology and Hydrochemistry of Salt Lakes and Groundwater Discharge Zones (Playa Lakes)

Previous investigations of salt lakes and groundwater discharge zones have focused mainly on their roles in regional water budgets (evaporation rate estimations), the leakage of brines back into regional groundwater systems and the geochemical evolution of their surface and subsurface brines. This section provides an overview of this work, and various aspects will be subsequently discussed in more detail in the relevant chapters.

1.5.1 Evaporation Rate Estimations

The fact that playa lakes accumulate large quantities of salt and are often points of regional groundwater discharge led to an early interest in quantifying both their hydrologic and solute budgets. Estimation of groundwater evaporation rates using physical methods was problematic in salt lake environments due to the harsh conditions and low evaporation rates (Tyler et al., 1997). Methods using open-water evaporation pans (Lee, 1912) have now been replaced by lysimeters (Lopes, 1986; Cochran et al.; 1988), mass and energy balance techniques (e.g. Malek and Bingham, 1993) and pore water chloride and δ^2H vs depth profiles (Ullman, 1985; Allison and Barnes, 1985). The latter method, employing environmental tracers, has been applied at Raak Plain (Howes, 1998).

The use of δ^2H vs depth profiles to estimate groundwater evaporation rates originated from a study by Zimmerman et al. (1967), who showed that pore water isotope vs depth profiles formed by evaporation in a saturated medium have a predictable shape resulting from a balance between upward evaporative downward diffusive fluxes. Barnes and Allison (1983) and Allison and Barnes (1985) extended this work to chloride and δ^2H depth profiles in non-vegetated unsaturated soils, where water contents change with depth, estimating an evaporation rate of 63 mm/y from the "normally dry" surface of Lake Frome, Australia. Ullman (1985) also used chloride and bromide profiles to estimate evaporation rates of between 9 and 28 mm/y from the salt-crusted surface of Lake Eyre, South Australia.

1.5.2 Brine Leakage

Early physical and chemical studies of salt lakes (e.g. Langbein, 1961; Hahl et al., 1965; Eugster and Jones, 1979) assumed closed systems where there were no surface or subsurface water and solute outflows. However, more recent studies have emphasized the role of pore fluids in the storage of solutes, preservation of evaporites and leakage of brine back into regional groundwater systems (e.g. Jones and Van Denburgh, 1966; Lerman, 1979; Berner, 1980; Spencer et al., 1985 (a&b); Bowler, 1986). Bowler (1986) suggested that vertical transfer of large volumes of brine from a subsurface brine pool to the regional groundwater system occurs at Lake Frome, Australia, despite the presence of a relatively impermeable clay / sand layer. The subsurface brine pool at Lake Frome partially penetrates the aquifer, as do those at Owens Lake, California (Rogers, 1993) and the Smith Creek Valley playa, Nevada (Thomas et al., 1989).

In other cases, for example the Tyrrell Basin, Australia (Teller et al., 1982; Macumber, 1991, 1992), and Mono Lake, California (Rogers, 1993; Rogers et al., 1992), the brine body has completely filled the aquifer below the playa. The Tyrrell Basin will be discussed separately in Section 3.3, as it is the closest well-studied system of playa lakes in hydrogeological setting and brine chemistry to Raak Plain. At Mono Basin, snowmelt and rainfall, originating in the adjacent Sierra Nevada, enter Mono Lake (approx. 200 km^2) via streams and groundwater seepage and are evaporated. The resulting saline (> 18,000 ppm) groundwater plume extends 2 km below the lake to the bottom of the basin fill and accounts for 80% of the dissolved solutes in the basin (Rogers, 1991). However, Rogers (1993) used a $^{36}Cl/Cl$ mass balance model at Mono Basin to show that the current inventory of chloride, including the subsurface brine , does not account for inflows over the past 3 to 4 million years, indicating leakage of solutes out of the basin by some undetermined mechanism.

1.5.3 Salt Lake Geochemistry and Brine Evolution

The hydrochemical evolution of salt lake brines has also been the subject of rigorous study over the past 3 to 4 decades, and has developed in parallel with the understanding of salt lake hydraulics. As described above, the earliest studies of brine hydrochemistry assumed evapo-concentration of inflow in basins that were closed to both surface outflow and groundwater leakage (e.g. Garrels and Mackenzie, 1967; Hardie and Eugster, 1970). The resulting model for brine evolution was based on the fundamental principle of the chemical divide, discussed in more detail in Section 3.2. However, this was an over-simplification of real world systems and further work considered other mechanisms that can influence the final compositions of brines, including magnesium ion removal by dolomitization, sulfate reduction, ion exchange and adsorption and successive wetting and drying cycles (Drever and Smith, 1978; Eugster and Hardie, 1978; Eugster and Jones, 1979).

The Eugster-Jones-Hardie model (Eugster, 1970; Eugster and Hardie, 1978; Eugster and Jones, 1979) was based on the saline Na-CO_3-SO_4-Cl brines of the Lake Magadi Basin, in the East African Rift Valley, Kenya. Also studied extensively have been the Na-(Mg)-Cl and Na-(Mg)-Cl-(SO_4) brines of Great Salt Lake, Utah, a surface water lake fed by mountain runoff (Hahl and Langford, 1964; Hahl and Handy, 1969; Whelan, 1973; Spencer et al., 1985 (a&b)), Mono Basin, California, described above and the Tyrrell Basin, Australia (Herczeg and Lyons, 1991; Macumber, 1991, 1992; Long et al., 1992). These will be discussed in more detail in Sections 3.2 and 3.3.

1.5.4 The Influence of Leakage on the Chemical Evolution of Brines

The process of brine leakage described in Section 1.5.2 above has often been used to explain deficits in the solute budgets for terminal basins, the presence of non-equilibrium evaporite mineral assemblages, and the transport of brine plumes over large distances (e.g. Wood and Sanford, 1990; Barnes et al., 1990). Both Wood and Sanford (1990) and Chambers et al. (1995) used geochemical models to show that observed mineral assemblages in playa lakes are strongly dependent on the ratio of brine leakage to inflow.