



# Hydrological and geochemical processes controlling variations in $\text{Na}^+ - \text{Mg}^{2+} - \text{Cl}^- - \text{SO}_4^{2-}$ groundwater brines, south-eastern Australia

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## ARTICLE INFO

### Article history:

Received 6 November 2007

Received in revised form 31 January 2008

Accepted 2 February 2008

Editor: D. Rickard

### Keywords:

Brine chemistry

Evaporites

Salt lakes

Playa lakes

Groundwater discharge

## ABSTRACT

Evaporative discharge of regional groundwater from more than 50 playa lakes at the 400 km<sup>2</sup> Raak Plain groundwater discharge complex forms a very large reservoir of hypersaline  $\text{Na}^+ - \text{Mg}^{2+} - \text{Cl}^- - \text{SO}_4^{2-}$  type subsurface brine, but very little salt precipitates. The main processes controlling the formation and evolution of brine compositions are: (a) evapo-concentration of the regional groundwater, (b) mixing between the high- and low-TDS end-members and (c) subsurface gypsum precipitation. Uniformity in these processes across the entire groundwater discharge complex, coupled with the chemical similarity between the regional groundwater and local recharge sources, has led to remarkably similar brine chemistries and evaporite compositions across Raak Plain. The similarity between the source waters and the ultimate brine compositions indicates that solutes are generally conserved within the system. The greatest variation in groundwater chemistry at Raak Plain was found to occur within the playa lakes, particularly within the top 1–2 m of the subsurface brine body, rather than amongst the different lakes. This small-scale variability is a result of 1) advective and diffusive mixing between the hypersaline shallow (<3 m below ground surface) brines and the less saline underlying groundwaters, 2) the transient precipitation and re-dissolution of surface salt efflorescences, and 3) differential diffusion rates of the various solutes over long time scales (>20 kyr).

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## 1. Introduction

The geochemical evolution of continental brines and playa lakes are important components of palaeoclimatic studies and their understanding is also recognised as being integral to maintaining unique flora and fauna in fragile saline ecosystems (Bowler, 1986; Duktiewicz et al., 2000; Yechiechi and Wood, 2002). Furthermore, the study of these natural saline systems assists with defining the major processes leading to salinisation of land and water due to anthropogenic activities (Herczeg et al., 1992; Simmons et al., 2002). Many studies have focused on surface water dominated saline lakes with relatively less emphasis on the interactions between the surface systems and subsurface brines (e.g. Garrels and Mackenzie, 1967; Eugster and Jones, 1979).

Saline lakes that derive the major fraction of their solutes from surface water inflows often lead to  $\text{Na}^+ - \text{HCO}_3^-$  type brines (e.g. Garrels and Mackenzie, 1967; Eugster and Jones, 1979; Connell and Dreiss,

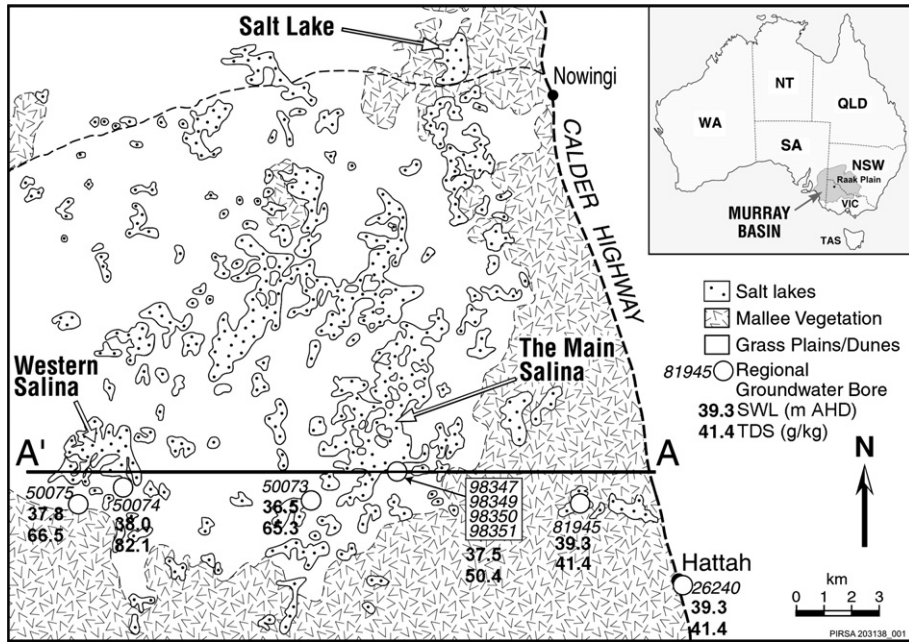
1995; Sinha and Raymahashay, 2004). Great Salt Lake, Utah on the other hand provides an example of a  $\text{Na}^+ - \text{Cl}^-$  brine (Spencer et al., 1985a,b), with inputs from hydrothermal  $\text{Na}^+ - \text{Cl}^-$  brines and mineral dissolution and diffusion process in the upper few metres. Large playa lakes and groundwater discharge complexes located throughout inland Australia are also characterized by substantial subsurface brine reservoirs, which are  $\text{Na}^+ - \text{Mg}^{2+} - \text{Cl}^- - \text{SO}_4^{2-}$  dominated (e.g. Jack, 1921; Draper and Jensen, 1976; Jacobson and Jankowski, 1989; Macumber, 1991; Herczeg and Lyons, 1991). These brines are thought to have evolved from an original marine aerosol signature of rainwater, concentrated by evapo-transpiration as water moves through the large, semi-arid groundwater basins (Chivas et al., 1991; Herczeg and Lyons, 1991; Mazor and George, 1992; Herczeg et al., 2001). Once this groundwater arrives at playa lakes at the basin centres, it has already reached Total Dissolved Solids (TDS) concentrations of 30–50 g/kg, and the geochemical processes that occur subsequently in these environments represent the late-stage evolution of a seawater-like source in a continental setting.

While the overall geochemical characteristics of the above-mentioned brine evolution model are observed in the field, there are a number of unanswered questions remaining. For example, subsurface groundwater salinities vary by a factor of 20 within the groundwater discharge zones, up to hypersaline levels, yet there is a general lack of preserved minerals at the playa surface. Our lack of

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**Fig. 1.** The Raak Plain groundwater discharge complex, located in the central Murray Basin of south-eastern Australia. Regional groundwater flows approximately east–west in the Parilla Sand aquifer below the discharge complex. The study was focused along the east–west transect, AA', although Salt Lake, in the north of the discharge complex, was also included.

understanding of the reasons for this suggests that physical and chemical processes at the scale of  $10^{-2}$  to  $10^2$  m are not well understood. This study takes advantage of a natural field setting in the central Murray Basin of south-eastern Australia, to examine at a range of scales the important physical and chemical processes affecting variability in brine composition and evaporite mineralogy within a large regional groundwater discharge complex. Specifically, we evaluate the relative importance of processes such as dissolution and re-precipitation of salts near the playa surfaces, diffusion and advective fluxes (recharge and discharge).

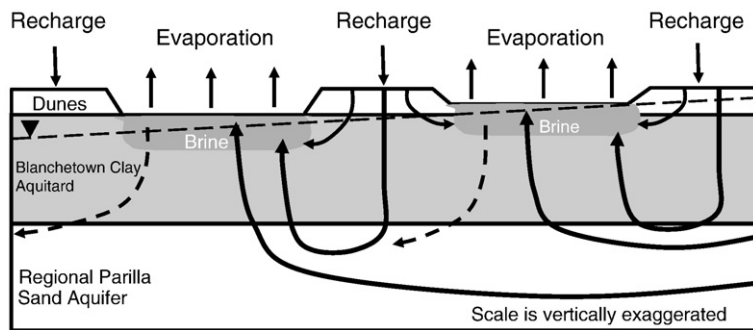
**2. Site description**

Groundwater flow in the flat, saucer-shaped Murray Basin of south-eastern Australia is focused radially inward towards the centre, where there are numerous active and “fossil” (currently inactive) groundwater discharge complexes, with associated brine bodies (Brown, 1989). Raak Plain is the largest (400 km<sup>2</sup>) of these groundwater discharge complexes and contains more than 50 small playa

lakes from which regional groundwater evaporates (Macumber, 1991) (Fig. 1). The predominant shallow aquifer feeding the discharge complexes in this region is the unconfined to semi-confined Pliocene Parilla Sand aquifer, a 60–70 m thick layer of unconsolidated to partially consolidated sands (Fig. 2). It is semi-confined below Raak Plain, being overlain by thicker deposits of the Plio-Pleistocene Blanchetown Clay aquitard, a sequence of gypsiferous clays, silts and minor fine sands (Fig. 2) (Rogers et al., 1995).

The climate in the central Murray Basin is semi-arid (rainfall ~250 mm/yr; potential evapo-transpiration ~2000 mm/yr), and groundwater in the Parilla Sand aquifer becomes progressively more saline towards the centre of the basin (up to 50 g/kg TDS; Brown, 1989; Evans and Kellett, 1989) due to the accession of salts from the evapo-concentration of rainfall (Jones et al., 1994; Herczeg et al., 2001). The residual solutes that accumulate beneath the playa lakes following evaporation of regional groundwater form hypersaline (>250 g/kg) groundwater brines (Fig. 2).

The landscape of the groundwater discharge complex consists of playa lakes intermingled with sandy plains, samphire-vegetated



**Fig. 2.** Conceptual diagram of the processes believed to contribute to the formation of the subsurface groundwater brines below the playa lakes at Raak Plain. Regional and locally recharged groundwater is evaporated from the surfaces of the playa lakes. The remaining solutes form the hypersaline (up to 250 g/kg) subsurface brines, which may be contained below the playa lakes by the Blanchetown Clay aquitard or eventually leak back into the regional groundwater system. The potentiometric surface (dashed line) shown is for the regional Parilla Sand aquifer.

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