



Differences in groundwater and chloride residence times in saline groundwater: The Barwon River Catchment of Southeast Australia



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ABSTRACT

The residence times of groundwater and chloride and the processes contributing to the development of saline (total dissolved solids (TDS) up to 45,379 mg/L) groundwater within the Barwon River Catchment of southeast Australia were investigated using major ion, stable isotope ($\delta^{18}\text{O}$, $\delta^2\text{H}$, and $\delta^{13}\text{C}$) and radioactive isotope (^3H , ^{14}C , ^{36}Cl) geochemistry. The elevated groundwater salinity in the region is primarily due to evapotranspiration and recycling of solutes in saline lakes with minor contributions from weathering of halite, silicate and calcite minerals. Groundwater residence times estimated from ^{14}C vary from modern to ~20 ka; for groundwater with lower ^{14}C activities, the estimated residence times vary significantly depending on the assumed flow model and the ^{14}C activity of recharge. Chloride residence times downgradient of Lake Murdeduke (a saline through-flow lake in the centre of the catchment) are greater than the corresponding groundwater residence times due to the recycling of Cl within the lake. Precise estimates of chloride residence time could not be determined using ^{36}Cl due to $R^{36}\text{Cl}$ in precipitation being lower than that of groundwater. This is most likely due to $R^{36}\text{Cl}$ values in rainfall having been higher in the past than they are at present due to climate variability. $\delta^{18}\text{O}$, $\delta^2\text{H}$, and $\delta^{13}\text{C}$ values also suggest that the region has experienced increasingly more evaporative conditions with time. The results of this study demonstrate that, while Cl is a useful tracer of hydrological processes, it must be applied carefully in arid and semi-arid regions of the world. In particular, recharge rates calculated using chloride mass balance may be underestimated where recycling of Cl has occurred.

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1. Introduction

Understanding groundwater systems requires being able to account for the behaviour of both the water and the solutes (Edmunds, 2009). Chloride is probably the most widely studied solute as it is one of the most abundant ions in groundwater and its concentration is readily measured. Because its concentration is not significantly impacted by biogeochemical reactions, ion exchange or decay, and because it forms minerals only at very high concentrations, chloride is commonly used to estimate groundwater recharge rates and residence times (e.g., Allison and Hughes, 1978; Wood and Sanford, 1995; Scanlon et al., 2006). Use of Cl in this way makes the explicit assumption that its residence time is equivalent to that of the groundwater in which it is contained. However, this assumption may not always hold true. For

example, in arid and semi-arid regions, Cl may be recycled over long timescales within saline lakes. Secondly, diffusion of old Cl from clays into adjacent aquifers may occur. Thirdly, if halite dissolution within an aquifer occurs, the halite represents a source of potentially very old Cl. Each of these scenarios results in Cl having a longer residence time than the water in which it is contained. If chloride residence times are significantly older than groundwater residence times, the use of Cl to estimate recharge rates or groundwater residence times will produce erroneous results. The implications of this are significant in terms of managing groundwater resources, protecting ecosystems, and assessing salinity impacts.

Chloride residence times can be estimated from chloride concentrations within the groundwater, knowledge of aquifer thickness and porosity, and an estimate of the amount of chloride delivered annually via rainfall. As an independent measure, chloride residence times can also be estimated using ^{36}Cl . With a half-life of 301,000 years, ^{36}Cl can be used to determine chloride residence times within the range of approximately 100 ka to 1 Ma (e.g. Bentley et al., 1986a; Phillips et al., 1986). Groundwater residence times up to approximately 35 ka are

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frequently estimated in regional aquifers using ^{14}C . While ^{14}C has been widely used as a tracer, most studies have not taken into account long-term variations in the ^{14}C of the water that recharges the aquifer nor processes such as dispersion, which can impact calculated residence times.

In this study, we use ^{14}C , ^{36}Cl , and ^3H in combination with major ion geochemistry and stable isotopes to understand the timescales and processes responsible for the development of saline groundwater within the Barwon River Catchment in southeast, Australia. In addition, we examine whether groundwater residence times and chloride residence times within the catchment are equivalent. While there have been a number of studies that have utilised both ^{36}Cl and ^{14}C (e.g. Bird et al., 1989; Love et al., 2000; Kulongoski et al., 2008; Plummer et al., 2012; Scheiber et al., 2015), this is the first application of these tracers that we know of that specifically addresses the potential differences between groundwater and chloride residence times. The outcomes of this study will help understand the timescales and processes by which chloride is stored and exported from similar catchments globally, and will better inform the use of Cl as a tracer in saline groundwater.

2. Setting

The Barwon River Catchment is located in southwestern Victoria, Australia. Comprising an area of approximately 3900 km² (DataSearch Victoria, 2015), the catchment is utilised primarily for grazing, cropping and forestry. Settlements within the catchment include the major population centres of Geelong and Ballarat, as well as numerous rural communities. This investigation focuses primarily on that portion of the catchment located in the vicinity of Winchelsea and Inverleigh (Fig. 1a).

The catchment has a temperate climate, with average annual rainfall ranging from approximately 430 mm near Inverleigh to approximately 1600 mm in the Otway Ranges (Department of Environment, Land, Water and Planning, Water Measurement Information System, WMIS, 2015). The majority of rainfall occurs during the austral winter months (July to September) and, during the summer months, potential evaporation exceeds precipitation (Bureau of Meteorology, BOM, 2016). The Barwon River is perennial (Department of Environment, Land, Water and Planning, Water Measurement Information System, WMIS, 2015) and flows northwards from the Otway Ranges, which reach a maximum elevation of approximately 650 m above sea level. There are a number of permanent and semi-permanent brackish to hyper-saline lakes within the study area (Tweed et al., 2011). The largest of these is Lake Murdeduke, which has an area of approximately 1,660 ha, a depth of approximately 5 m and a mineral ('salt') crust that periodically forms along its shores. From an analysis of regional groundwater flow paths, salt and water balances, and geochemistry, Coram (1996) concluded that Lake Murdeduke was a through-flow lake, with groundwater entering the lake along its west shore and discharging along its east shore.

The basement of the catchment comprises the early-Cretaceous Otway Group, which consists primarily of volcanogenic sandstone and mudstone with minor amounts of shale, siltstone, and coal (Tickell et al., 1991). The Otway Group is overlain by Tertiary sediments comprising (in ascending order) the Eastern View Formation, the Demons Bluff Formation, the Clifton Formation, the Gellibrand Marl and the Moorabool Viaduct Sand. The Tertiary sediments are in turn overlain by the Quaternary Newer Volcanics, which crop out over approximately 40% of the catchment. This study is based on groundwater sampled primarily from the Newer Volcanics and the Moorabool Viaduct Sand (Fig. 1a).

The Moorabool Viaduct Sand is a thin (generally less than 10 m thick), discontinuous unit of clayey sand that crops out at various locations across the catchment, including to the west of Lake Murdeduke and near Winchelsea and Inverleigh. Coram (1996) estimated that the

hydraulic conductivity of this unit was approximately 4.3 m/day. The Newer Volcanics have been broadly grouped into "older-" and "younger-phase" deposits. The older-phase Newer Volcanics, which are comprised mainly of weathered olivine basalt, are considered to be a poor aquifer, with groundwater generally only occurring within joints, vesicles, and poorly-connected interflow layers. Gill (1989) estimated that the older-phase Newer Volcanics have an average transmissivity of approximately 200 m²/day (which assuming an average thickness of 15 m equates to a hydraulic conductivity of 13.3 m/day). The younger-phase Newer Volcanics consist of basalt, scoria, undifferentiated pyroclastics, pyroclastic surge deposits and pyroclastic fall deposits (Tickell et al., 1991). They are less weathered and have a higher degree of fracturing than the older phase volcanics. Coram (1996) estimated that the younger-phase Newer Volcanics have an average hydraulic conductivity of 8.6 m/day. Together, the older- and younger-phase Newer Volcanics have an average thickness of approximately 30 m in the study area. Also present are Quaternary lunette deposits along the east shore of Lake Murdeduke and alluvium along the Barwon River (Fig. 1a).

Regional groundwater in the lower Tertiary units flows from recharge areas in the Otway Ranges to the north or northeast (Witebsky et al., 1995; Petrides and Cartwright, 2006). Flow direction within the Newer Volcanics and underlying Moorabool Viaduct Sand is more variable and is influenced by local topography. Recharge occurs across the basin and groundwater discharges into the Barwon River (Cartwright et al., 2013a). Shallow groundwater in the headwaters of the catchment generally has low (<500 mg/L) total dissolved solids (TDS) concentrations but in the centre of the catchment, near Lake Murdeduke, TDS concentrations in excess of 10,000 mg/L are common (Coram, 1996). TDS concentrations as high as 1200 mg/L have also been reported within the Barwon River (Cartwright et al., 2013a), which is interpreted to be the result of highly saline groundwater inflow (Roderick, 1988; Gill, 1989).

3. Methods

3.1. Sampling

Samples of precipitation, groundwater, Lake Murdeduke lake water and Lake Murdeduke salt crust were collected from the study area between September 2014 and November 2015. The EC, pH and temperature of the water samples were measured in the field using a calibrated, TPS® hand-held water quality meter and probes and alkalinity was determined using a Hach® digital titrator and reagents. Sampling locations are shown in Figs. 1a, 1b and 2.

Precipitation samples were collected using rainfall collectors located along the east shore of Lake Murdeduke, on the east side of the West Barwon Reservoir and at Birnam in the Otway Ranges (Fig. 1a). Precipitation collectors consisted of polyethylene storage containers equipped with funnels positioned approximately one-half metre above ground level. Water within the containers was periodically sampled, the containers rinsed with ultrapure water and then replaced. Precipitation samples were collected on six occasions at Birnam, on two occasions at the West Barwon Reservoir and on one occasion at Lake Murdeduke.

Groundwater samples were collected in March 2015 from 22 groundwater monitoring bores that comprise part of the Victorian State Observation Bore Network (SOBN). All but three of the bores are located within the vicinity of Lake Murdeduke; the remaining bores are located approximately 20 to 30 km to the north of the lake (Fig. 1a). The sampled bores are screened within the Newer Volcanics, the Moorabool Viaduct Sand or lunette deposits at depths ranging from approximately 8 to 65 m below ground level. At several locations, adjacent bores are screened at different depths within a single geologic unit and/or between units. Bore details are summarised in Table 1. Standing water levels were measured prior to sampling using an electronic water level tape. Groundwater

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