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Discussions

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Using ^{14}C and ^{3}H to understand groundwater flow and recharge in an aquifer window

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Abstract

Knowledge of groundwater residence times and recharge locations are vital to the sustainable management of groundwater resources. Here we investigate groundwater residence times and patterns of recharge in the Gellibrand Valley, southeast Australia, where outcropping aquifer sediments of the Eastern View Formation form an "aquifer window" that may receive diffuse recharge and recharge from the Gellibrand River. To determine recharge patterns and groundwater flowpaths, environmental isotopes (³H, ¹⁴C, δ^{13}C, δ^{18}O, δ^{2}H) are used in conjunction with groundwater geochemistry and continuous monitoring of groundwater elevation and electrical conductivity. Despite the water table fluctuating by 0.9–3.7 m annually producing estimated recharge rates of 90 and 372 mm yr^{-1}, residence times of shallow (11–29 m) groundwater determined by ¹⁴C ages are between 100 and 10 000 years. ³H activities are negligible in most of the groundwater and groundwater electrical conductivity in individual areas remains constant over the period of study. Although diffuse local recharge is evident, the depth to which it penetrates is limited to the upper 10 m of the aquifer. Rather, groundwater in the Gellibrand Valley predominantly originates from the regional recharge zone, the Barongarook High, and acts as a regional discharge zone where upward head gradients are maintained annually, limiting local recharge. Additionally, the Gellibrand River does not recharge the surrounding groundwater and has limited bank storage. ¹⁴C ages and Cl concentrations are well correlated and Cl concentrations may be used to provide a first-order estimate of groundwater residence times. Progressively lower chloride concentrations from 10 000 years BP to the present day are interpreted to indicate an increase in recharge rates on the Barongarook High.

1 Introduction

Groundwater residence time can be defined as the period of time elapsed since the infiltration of a given volume of water (Campana and Simpson, 1984), or perhaps more

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accurately, the mean time that a mixture of waters of different ages have resided in an aquifer (Bethke and Johnson, 2008). The residence time of water within an aquifer is a key parameter in describing catchment storage and may be used to estimate historical recharge rates (Le Gal La Salle et al., 2001; Cook et al., 2002; Cartwright and Morgenstern, 2012; Zhai et al., 2013), elucidate groundwater flowpaths (Gardner et al., 2011; Smerdon et al., 2012), calibrate hydraulic models (Mazor and Nativ, 1992; Reilly et al., 1994; Post et al., 2013) and characterize the rate of contaminant spreading (Böhlke and Denver, 1995; Tesoriero et al., 2005). From a water resource perspective, information on groundwater residence times is required for sustainable aquifer management by identifying the risk posed to groundwater reserves against over-exploitation (Foster and Chilton, 2003), climate change (Manning et al., 2012) and contamination (Böhlke, 2002).

Unconfined aquifers may be recharged over broad regions leading to young groundwater at shallow depths (Cendón et al., 2014). On the other hand, the residence time of groundwater in confined aquifers generally increases away from discrete recharge areas. The geology of catchments is often complex and heterogeneous and, where aquifer material is exposed in more than one location, this offers a potential "window" for groundwater recharge (Meredith et al., 2012). It is important to document groundwater flow in such aquifer windows. If they act as recharge areas, changes in land-use such as agricultural development may introduce contaminants to the deeper regional groundwater systems. By contrast, if they are local discharge areas, use of regional groundwater may impact rivers, lake or wetlands that are receiving groundwater.

High river flows may also recharge shallow groundwater if the hydraulic gradient between the river and the groundwater is reversed during high flows (Doble et al., 2012). Episodic recharge of aquifers by large over-bank floods is also locally important (Moench and Barlow, 2000; Cendón et al., 2010; Doble et al., 2012), particularly in arid areas (Shentsis and Rosenthal, 2003); however, the potential for over-bank events to recharge aquifers in temperate areas is still poorly understood. Additionally, during high

flow, water from rivers is likely stored temporally in the banks (McCallum et al., 2010; Unland et al., 2014); however, the depth and lateral extent to which bank exchange water infiltrates the aquifer is not well documented. Understanding the capacity of rivers to recharge regional groundwater is important in understanding exchange within the hydrological cycle (Stichler et al., 1986; Chen and Chen, 2003). Furthermore, where surface-waters transport contaminants and have the potential to recharge the surrounding aquifer, this may lead to contamination and degradation of groundwater quality (Newsom and Wilson, 1988; Stuyfzand, 1989). Lastly, knowledge of residences times of groundwater in close proximity to the river can provide important information on groundwater-river interactions (Gardner et al., 2011). Local groundwater flowpaths in connection with rivers are often underlain by deeper regional flowpaths (Tóth, 1963) however the role these flowpaths play in contributing to river baseflow remains unclear (Sklash and Farvolden, 1979; McDonnell, 2010; Frisbee and Wilson, 2013; Goderniaux et al., 2013). This may be elucidated from understanding residence times of near-river groundwater (Smerdon et al., 2012).

1.1 Radiogenic tracers

Radioactive environmental isotopes, in particular ^{14}C and ^{3}H have proven useful tools for determining groundwater residence times (Vogel, 1974; Wigley, 1975). Produced in the atmosphere via the interaction of N_2 with cosmic rays, ^{14}C has a half life of 5730 years and can be used to trace groundwater with residence times up to 30 ka. The use of ^{14}C in dating groundwater was first discussed by Muennich (1957), and has subsequently been widely used due to the ubiquitous presence of dissolved inorganic carbon (DIC) in groundwater (Cartwright et al., 2012; Samborska et al., 2012; Stewart, 2012). The calculation of ^{14}C ages may be complicated if groundwater DIC is derived from a mixture of sources (Clark and Fritz, 1997). Where a large proportion of DIC is derived from the dissolution of ^{14}C-free carbonate minerals in the aquifer matrix, the ^{14}C originating from the atmosphere or soil zone will be significantly diluted. Additionally, geogenic CO_2 and CO_2 generated by the breakdown of organic matter

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during methanogenesis may provide additional sources of ^{14}C-free DIC. Groundwaters recharged post 1950 may have anomalously high ^{14}C activities (a^{14}C) due to the ^{14}C produced during atmospheric nuclear tests. Objective ^{14}C dating requires recognition and quantification of these processes. A number of models based on both major ion and stable C isotope geochemistry have been proposed to correct apparent ^{14}C ages (Han and Plummer, 2013).

   With a significantly shorter half-life (12.33 years), ^{3}H can be used to date groundwater with residence times of up to 100 years (Vogel et al., 1974). With the decay of the 1960s ^{3}H bomb-pulse peak in the Southern Hemisphere to near background levels unique ages may now be determined from single ^{3}H measurements (Morgenstern et al., 2010). As ^{3}H is part of the water molecule, there is negligible change to ^{3}H activities other than decay, providing an excellent tracer for the movement of water through hydrological systems (Michel, 2004). Used in conjunction with ^{14}C data, ^{3}H may also be used to study mixing in shallow aquifers (Le Gal La Salle, 2001; Cartwright and Morgenstern, 2012).

2 Study site

The Otway Basin is located in southwest Victoria, covering an area of 150 000 km^2. The basin was formed during the Cretaceous rifting of Australia and Antarctica (Briguglio et al., 2013) and is infilled with Upper Cretaceous and Cenozoic siliciclastic and calcareous sediments that form several aquifers and aquitards. The basin is divided into a number of sub-basins with regional groundwater flow paths originating at topographic highs. The Gellibrand catchment is one of these sub-basins. This study focuses on a 250 km^2 upland area of the catchment, the Gellibrand River Valley, which lies at the foothills of the Otway Ranges, directly south of the Barongarook High, a regional recharge zone, from which groundwater flows southwest along the Gellibrand River Catchment as well as eastward into the Barwon Downs Graben (Fig. 1).

Cretaceous Otway Group sediments of the Eumeralla Formation form the basement of the catchment and crop out in areas of higher relief. The Eumeralla Formation consists of thickly bedded siltstone, mudstone and volcanolithic sandstone. It has a low primary porosity and hydraulic conductivity and acts as a poor aquifer (Lakey and Leonard, 1982). Cenozoic sediments of the Wangerrip group overlie the bedrock and form major aquifers in the region to which groundwater flow is constrained (Van den Berg, 2009). The primary aquifer in the study area is the Eastern View Formation or equivalent Dilwyn Formation (Van den Berg, 2009; Petrides and Cartwright, 2006; Atkinson et al., 2013), composed of gravel, fine to coarse grained sand and major clays. The Eastern View Formation comprises predominantly quartz, feldspars and carbonates ($< 2\%$) and has hydraulic conductivities of 10^{-2} to 10^{2} m d^{-1} (Hortle et al., 2011). The Eastern View Formation is underlain by another productive aquifer, the Pebble Point Formation, however this is much thinner and is separated from the above layers by the Pember Mudstone. To the north the Eastern View Formation is confined by the Gellibrand Marl, a regional aquitard, which comprises 100–200 m of clay, and fine-grained silts of the Demons Bluff formation. Basaltic intrusions of the Quaternary Newer Volcanics are also present. The floodplain is covered with recent alluvial deposits of sand and clay.

The Gellibrand Valley contains a mixture of cool temperate rainforest on the valley sides and cleared agricultural pasture through which the Gellibrand River flows. Rainfall across the catchment averages ~ 1000 mm yr^{-1}, with the majority of rainfall falling in the Australian winter between June and September (Bureau of Meteorology, 2013). The Gellibrand River is gaining and groundwater contributes between 10 and 50 % to total river flow dependent on flow conditions (Atkinson et al., 2013). River flows are between 5×10^{4} m^{3} day^{-1} to 2×10^{6} m^{3} day^{-1} (Fig. 2c), with low flows during summer months (December to March) and high flows and flooding during winter (June–August) (Victorian Water Resources Data Warehouse, 2013), during which, there is the potential for aquifer recharge from overbank flow and bank storage. Regional recharge occurs on the Barongarook High where the Eastern View Formation crops out, however

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there is also potential for localised recharge within the Gellibrand River Valley, as the Eastern View Formation crops out there.

Though groundwater residence times in the Otway Basin have been explored in the Gambier Embayment (Love et al., 1994) and nearby Barwon River catchment (Petrides and Cartwright, 2006), little is known of the residence times of groundwater in the Gellibrand River catchment. Here we evaluate groundwater residence times in the upper Gellibrand River Catchment, where the Eastern View Formation is exposed and regular episodic river floods occur, to identify whether groundwater recharge occurs in this part of the groundwater system. This is important in understanding the potential impacts of landuse change and pollution in the catchment as well as understanding the dynamics of recharge in catchments where aquifer material is exposed in more than one location. Radioactive tracers ^{14}C and ^{3}H are used to determine residence times and define groundwater flow paths whilst major ion chemistry is employed to determine dominant geochemical processes. Water table fluctuations and groundwater electrical conductivities are also continuously monitored. These easily measurable, robust parameters can be used to observe changes in storage and infer sources of aquifer recharge (Vogt et al., 2010) and allow for comparison with radioisotopes in understanding the dynamics of groundwater systems.

3 Methods

A number of groundwater monitoring bores which form part of the Victorian State Observation Bore networks are present in the Gellibrand Valley (Victorian Water Resources Data Warehouse, 2013). These are screened in the Eastern View Formation, with depths of between 0 and 42 m. Bores located within 25 m from the Gellibrand River generally have screen depths between 11 and 15 m, whilst bores located on the flood plain have depths between 21 and 42 m. Groundwater from the Eastern View Formation was sampled from 13 bores. 10 of these are located within 25 m from the river in a 14 km^2 area of the catchment, with 3 further samples

taken from bores situated on the flood plain between 1 and 2 km from the river. Groundwater was sampled using an impeller pump set in the screen with 2 to 3 bore volumes purged before sampling. Electrical conductivity (EC) and pH of groundwater were measured in the field using a calibrated TPS WP-81 conductivity/pH meter and probes. To assess transient changes in groundwater levels and EC, Aqua Troll 200 (in situ) data loggers were deployed in June 2011. A significant drop in EC in near-river groundwater is shown in some bores following flooding in June 2012 when bores were overtopped. However immediately upon pumping in October 2012 (B108934, B108940) and April 2013 (B108916), the EC of the groundwater returned to pre-flood EC values. We interpret this as floodwater that infiltrated down the bore which was not displaced by groundwater prior to pumping, and these data have been omitted. Rainfall samples were also collected in the catchment throughout the study period for chemical analysis.

Cations were analysed on a Thermo Finnigan X Series II Quadrupole ICP-MS on samples that had been filtered through 0.45 µm cellulose nitrate filters and acidified to pH < 2. Anions were measured on filtered unacidified samples using a Metrohm ion chromatograph. The precision of major ion concentrations based on replicate analyses is ±2 %. Charge balances are within ±5 %. Stable isotope ratios were measured using Finnigan MAT 252 and ThermoFinnigan DeltaPlus Advantage mass spectrometers. δ^{18}O values were measured via equilibration with He-CO$_2$ at 32 °C for 24–48 h in a Finnigan MAT Gas Bench whilst δ^2H values were measured by the reaction of water samples with Cr at 850 °C using a Finnigan MAT H/Device. Both δ^{18}O and δ^2H were measured against an internal standard that has been calibrated using the IAEA, SMOW, GISP and SLAP standards. Data was normalised following methods outlined by Coplen (1988) and are expressed relative to V-SMOW where δ^{18}O and δ^2H values of SLAP are −55.5 and −428 ‰ respectively. Precision is ±1 ‰ for δ^2H and ±0.2 ‰ for δ^{18}O.

^{14}C and ^{3}H samples of groundwater were measured at the Australian Nuclear Science and Technology Organisation (ANSTO) and the Tritium and Water Dating

Laboratory, Institute of Geological and Nuclear Sciences (GNS), (New Zealand). For ^{14}C analysis performed at ANSTO, CO_2 was extracted from water samples in a vacuum line using orthophosphoric acid and converted to graphite through reduction with excess H_2 gas in the presence of an iron catalyst at 600 °C. ^{14}C concentrations were measured using a 10 kV tandem accelerator mass spectrometer. δ^{13}C values for these samples are derived from the graphite fraction used for radiocarbon via EA-IRMS.

For ^{14}C samples measured at GNS, CO_2 was extracted from groundwater samples through addition of orthophosphoric acid. CO_2 was made into a graphite target and analysed by AMS. An aliquot of the extracted CO_2 was used for δ^{13}C analysis. ^{14}C activities are expressed as pMC (percent modern carbon) where pMC = 100 % corresponds to 95 % of the ^{14}C concentration of NBS oxalic acid standard (Stuiver and Polach, 1977), with a precision of ^{14}C/^{12}C ratios of ±0.5 (Fink et al., 2004). At both ANSTO and GNS, samples for ^{3}H were distilled and electrolytically enriched prior to being analysed by liquid scintillation counting as described by Neklapilova et al. (2008a, b) and Morgenstern and Taylor (2009). ^{3}H activities are expressed in Tritium Units (TU) with a relative uncertainty of ±5 % and a quantification limit of 0.13–0.14 TU at ANSTO and 0.02 TU and a relative uncertainty of 2 % at GNS.

4 Results

4.1 Groundwater elevations

Groundwater elevations decrease from 230 m relative to the Australian Height Datum (AHD) on the Barongarook High to < 60 mAHD within the Gellibrand Valley (Fig. 1), with groundwater flowing from the Barongarook High towards the Gellibrand Valley and then westward. Groundwater elevations from all depths and positions within the Gellibrand Valley are in phase and fluctuate between 1 and 3 m annually (Fig. 2a). Rising water tables follow winter rainfall between June and August (Fig. 2c) and head gradients at nested sites are upwards (Fig. 2b). The Gellibrand River has high water levels

that result in flooding during winter months (June–August) and low flows in summer (December–March) (Fig. 2c).

4.2 Groundwater geochemistry

The chemistry of groundwater in the Gellibrand Valley is summarised in Table 1. Groundwater has electrical conductivities between 140 and $600\,\mu S\,cm^{-1}$ and pH ranging from 4.8 to 6.0. Groundwater from close proximity to the river generally has higher EC values (144–$545\,\mu S\,cm^{-1}$) than groundwater further back on the floodplain (149–$220\,\mu S\,cm^{-1}$). The major ion chemistry of groundwater is similar across the catchment, and the groundwater is Na-Cl type. Cl constitutes between 68 and 92 % of total anions on a molar basis, with HCO_3 accounting for 0–25 %. Increases in Cl concentrations are associated with a decrease in HCO_3. Na comprises between 60 and 85 % of total cations with Ca constituting 1–10 %, Mg constituting 0–10 % and K constituting 0–10 %. Increased Na concentrations are associated with decreases in both Ca and Mg concentrations. Molar Cl/Br ratios are between 400 and 600 and do not increase with increasing Cl (Fig. 3b), molar Na/Cl ratios are 0.7–1.3 and also remain stable with increasing Cl concentrations (Fig. 3a). Both Cl/Br and Na/Cl ratios of groundwater samples are similar to those measured in rainfall in southeast Australia (Blackburn and Mcleod, 1983). There is a weak correlation between Ca and HCO_3 (Fig. 3c) and between Ca and O_4 (Fig. 3d).

4.3 Stable Isotopes (δ^2H, δ^{18}O, δ^{13}C)

δ^{18}O and δ^2H values of groundwater define a narrow field (δ^{18}O = −4 to −6‰ and δ^2H = −30 to −40‰) that is close to both the global and local meteoric water lines (Fig. 4). The Gellibrand Valley is located between Melbourne and Adelaide, with groundwater generally plotting between the average isotopic compositions of meteoric waters located in these areas. δ^{13}C values of DIC from groundwater range from −19.8 to −25‰, with an average of 21.7‰ (Table 1).

4.4 ^{13}C, a^{14}C and ^{3}H concentrations

The a^{14}C of groundwater ranges from 29 to 101.5 pMC. ^{3}H activities are below detection for the majority of groundwater samples (Table 1), with the exception of bores 80 732, 80 735 and 110 737 which have activities of 1.02, 1.47 and 1.24 TU, respectively. Groundwater from these bores has a^{14}C > 90 pMC. The distribution of a^{14}C and ^{3}H values across the catchment is heterogeneous with no relationship to depth or along lateral groundwater flowpaths. A strong inverse correlation (R^2 = 0.87) is observed with Cl values. High a^{14}C groundwater is associated with low Cl concentrations, with the decrease in a^{14}C through radioactive decay matched by an enrichment of chloride ions. A similar correlation is also observed for Na (R^2 = 0.855), K (R^2 = 0.82), Ca (R^2 = 0.6) and Mg (R^2 = 0.54).

4.5 Continuous electrical conductivity

Continuous groundwater EC records for a number of near-river bores (excluding B110737, situated on the flood-plain) are shown in conjunction with changes in river height for the study period (Fig. 6). Groundwater EC in all bores for the majority of the dataset show little or no response to changes in river height. Minor changes in EC correlate to sampling events in which groundwater bores were pumped.

5 Discussion

5.1 Groundwater chemistry

Understanding geochemical processes in groundwater is required for correction of ^{14}C ages and in documenting groundwater flow and recharge. Processes which govern the evolution of groundwater geochemistry and sources of solutes in the Eastern View Formation can be determined from the major ion geochemistry. The observation that Cl/Br ratios are between 500 and 1000, which is similar to those expected in rainfall,

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and do not increase with increased TDS implies that evapotranspiration rather than halite dissolution is the major process controlling groundwater salinity (Herczeg et al., 2001; Cartwright et al., 2006). This conclusion is also consistent with an absence of halite in the aquifer lithologies. The δ^{18}O and δ^2H values of groundwater do not define evaporation trends, implying that transpiration in the soil zone or upper parts of the aquifer is likely to be more dominant over evaporation. Na/Cl ratios in groundwater are also similar to those in local rainfall (~ 1.00) implying that silicate weathering is limited (Edmunds et al., 2002), whilst the increase in Na concentrations at the expense of Ca may indicate ion exchange reactions on the surface of clay minerals. That Ca and mHCO$_3$ are poorly correlated suggests that only negligible dissolution of calcite has occurred. A handful of groundwater samples have a $1:1$ Ca : SO$_4$ ratio indicating some minor gypsum dissolution may take place. Together, the major ion geochemistry suggests that water-rock interaction is limited with minimal silicate weathering, negligible dissolution of halite and carbonate minerals and some minor dissolution of gypsum. As is the case elsewhere in southeast Australia, including within the Otway basin, the primary geochemical process is evapotranspiration promoted by the moderate rainfall and water-efficient native vegetation, and the groundwater salinity is largely controlled by the degree of evapotranspiration during recharge (Herczeg et al., 2001; Bennetts et al., 2006; Petrides and Cartwright, 2006).

5.2 Water table fluctuations

Annual cycles of groundwater elevations are present in all groundwater bores, which were screened 11–40 m below the ground surface. Fluctuations in groundwater levels across the Gellibrand River Valley are likely to reflect changes to the water table in response to rechargeThe magnitude of annual water table fluctuations recorded in data loggers is similar to those over the previous 30 years (Fig. 6). Recharge was estimated for years 2012 and 2013 using the water-table fluctuation

method Eq. (1):

$$R = S_y \cdot \Delta h / \Delta t \tag{1}$$

(Scanlon et al., 2002), where S_y is specific yield, Δh is the change in water table height between the hydrograph recession and hydrograph peak and Δt is time. The water table rise is estimated as the difference between peak groundwater levels and the extrapolated antecedent recession. The estimate of recharge from this method is sensitive to the estimate of the specific yield. S_y is assumed to be 0.1 which is close to the measured effective porosity of the Eastern View Formation (Love et al., 1993), and takes into account the presence of finer sized sediments such as silt and clay in the aquifer. Annual water table fluctuations are between 0.9 and 3.7 m across all bores, which for S_y values of 0.1, imply that $R = 130$ to $372\,\mathrm{mm\,yr^{-1}}$ in 2012 (mean of $200\,\mathrm{mm\,yr^{-1}}$) and 90–$300\,\mathrm{mm\,yr^{-1}}$ in 2013 (mean of $164\,\mathrm{mm\,yr^{-1}}$). This equates to between 11 and 32 % of rainfall in 2012 and 12 and 28 % of rainfall in 2013. The bores are screened 11.2–42 m below the ground surface and thus these recharge estimates will be minima due to the attenuation of pressure variations with depth (Scanlon et al., 2002). Recharge estimates are also susceptible to the value of specific yield, particularly where the aquifer is composed of finer sized sediments such as silt and clay. Regardless, estimates using bore hydrographs indicate that significant groundwater recharge to the unconfined Eastern View aquifer in the valley occurs via diffuse recharge.

5.3 ^{14}C ages

The groundwater in the Eastern View Formation is not anoxic (Victorian Water Resources Data Warehouse, 2013), nor are there coal seams hence methanogenesis is unlikely to be a source of DIC. Likewise there are no obvious sources of geogenic CO_2 in this area. Based on the major ion geochemistry, only minor calcite dissolution occurs in the Eastern View Formation, which is to be expected as the Cenozoic aquifers

are siliceous and contain only minor carbonate minerals. While only minor carbonate dissolution is likely, determination of groundwater residence times requires this to be taken into account. If it is assumed that closed system dissolution of calcite in the aquifers is the major process, the fraction of C derived from the soil zone (q) may be derived from the $\delta^{13}C$ values of DIC ($\delta^{13}C_{DIC}$), carbonate ($\delta^{13}C_{cc}$) and recharging water ($\delta^{13}C_r$) via Eq. (2):

$$q = \frac{\delta^{13}C_{DIC} - \delta^{13}C_{cc}}{\delta^{13}C_r - \delta^{13}C_{cc}} \tag{2}$$

(Clark and Fritz, 1997). The calcite is assumed to have a $\delta^{13}C$ of ∼ 0‰ (Love et al., 1994; Petrides and Cartwright, 2006) as is appropriate for marine sediments. $\delta^{13}C_r$ is calculated from the $\delta^{13}C$ of the soil carbon in the recharge zone. Pre-land clearing vegetation in southeast Australia was dominated by eucalypts that have $\delta^{13}C$ values of −30 to −27‰ (Quade et al., 1995). Assuming a ∼ 4‰ ^{13}C fractionation during outgassing (Cerling et al., 1991), $\delta^{13}C$ values of soil CO_2 would be −26 to −23‰ (average of −24.5‰). At 20°C and pH 6.5, $\delta^{13}C_r$ calculated from the fractionation data of Vogel et al. (1970) and Mook et al. (1974) is ∼ −20‰. Although the calculated $\delta^{13}C_r$ values require the pH and temperature of recharge and the $\delta^{13}C$ of the soil zone CO_2 to be estimated, they are similar to those from other studies in southeast Australia and consistent with the predicted $\delta^{13}C$ values of DIC in equilibrium with calcite in the regolith (Quade et al., 1995; Cartwright, 2010). Calculated q values are between 0.85 and 0.97 (Table 2), implying that only 10–15 % of DIC in groundwater from the Eastern View formation is derived from calcite in the aquifer, this is similar to the expected contribution of calcite dissolution in siliceous aquifers (Vogel et al., 1970) and similar to other estimates from the Otway Basin (Love et al., 1994; Petrides and Cartwright, 2006).

Using the q values from Table 2, ^{14}C ages (t) corrected for closed-system calcite dissolution are calculated from Eq. (3); where $a^{14}C$ is the activity of ^{14}C in groundwater

DIC, and $a_o{}^{14}C$ is the activity during recharge (assumed to be 100 pM C).

$$t = -8376 \ln \left(\frac{a^{14}C}{q \cdot a_o{}^{14}C} \right) \qquad (3)$$

Radiocarbon ages for groundwater in the Eastern View Formation range from 380 to 9260 years (Table 2) with the exception of bores B110737, B80732, B80735 which have $a^{14}C > 100$ pM C and represent groundwater that has a large component of water recharged during or after the atmospheric nuclear tests in the 1950s to 1960s. The majority of ^{14}C ages however, suggest that groundwater in the valley has long residence times (Fig. 7).

5.4 ^3H activities and recharge rates

With a shorter half-life, ^3H activities can infer the presence of modern groundwater. The water table fluctuations imply that the Gellibrand Valley receives considerable recharge year (90–370 mm yr^{-1}). Although head gradients at nested sites are upwards implying that the valley is a groundwater discharge zone (Fig. 2b), these may be reversed during periods of high rainfall. If local recharge is significant in recharging the groundwater system across the valley, it would be expected that the groundwater would have relatively high ^3H activities. Recently-recharged groundwater in other Victorian catchments has ^3H activities up to 3.6 TU (Cartwright and Morgenstern, 2012).

^3H activities in most of the groundwater from the Gellibrand Valley are negligible. Much of this groundwater is from within 5–10 m of the water table, suggesting that any recharge penetrates only to a limited depth, and does not mix with the bulk of the water in the Eastern View Formation. The exception to this is groundwater from the southern edge of the valley where the Eastern View Formation overlies the basement rock (Eumeralla Formation). Here both ^3H activities and ^{14}C activities are higher implying that recharge to the deeper parts of the aquifer locally occurs.

The Gellibrand River has the potential to recharge regional groundwater during high river stages and episodic floods. Aquifer recharge from surface water can be assessed by combining data from groundwater EC values and ^{3}H activities. The EC of river water varies between 120 and 200 μS cm^{-1} and is lower than that of groundwater in the catchment throughout the year. ^{3}H activities of river water are between 1.24 and 2.0 TU during baseflow conditions (Atkinson et al., 2013), and may be higher during high flow events as local modern rainfall, which is likely to comprise a significant component of river flow at thoese times, has activities of 2.4–3.2 TU (Tadros et al., 2014). Significant amounts of aquifer recharge through overbank events or bank exchange should result in groundwater with low EC values, and high ^{3}H activities near the river.

Except for in June 2012 when the bores were overtopped, groundwater EC was constant throughout the study period and there is no inverse relationship to river height (Fig. 6). This indicates there is little exchange of river water to the depth of the aquifer sampled by the bores. Additionally the activities of ^{3}H in near-river bores are negligible, again suggesting that recharge from the river does not penetrate more than a few metres into the adjacent aquifer. Thus, flow through the river bank or river flooding does not appear to be a significant mechanism of recharge in the Gellibrand Valley.

5.5 Groundwater flowpaths and conceptual model

Radiocarbon ages are up to 10 ka implying that the groundwater in the Gellibrand Valley has a long residence time; in turn this implies that the area is a regional discharge zone. Most of the groundwater originates on the Barongarook High, and this region potentially provides a substantial proportion of baseflow to the Gellibrand River. The large range of ^{14}C ages in the valley is a likely result of heterogeneous geology, where the presence of low hydraulic conductivity sediments such as silt and clays in the Eastern View formation lead to variable velocities along groundwater flowpaths. Groundwater travel times may also be determined using the present day hydraulic gradients. From Darcy's law and assuming a porosity of 0.1 (Love et al., 1994) and a hydraulic conductivity of 0.2–2 m day^{-1} (Love et al., 1993) calculated travel times are between 1000 and

10 000 years, which are similar to those implied by the ^{14}C ages. This supports the idea that groundwater in the valley is predominantly regional groundwater derived by recharge on the Barongarook High. The high ^{3}H activities in groundwater bores situated away from the river imply local recharge in that area to depths of 21–42 m.

However for the most-part, shallow groundwater in the Gellibrand valley, including in the near-river environment is predominantly regional groundwater. Though groundwater elevations display clear annual cycles and winter months are punctuated by high river flow, localised recharge from both of these processes combined is stored in the upper < 10 m of the aquifer. The presence of silts and clays on the floodplain and riverbanks combined with strong upwards hydraulic gradients in the Eastern View Formation, driven by groundwater flow from the Barongarook High, ensure that recharge in the near-river environment does not penetrate deep within the aquifer (Fig. 8).

5.6 ^{14}C ages and Cl

The good correlation of $a^{14}C$ with chloride implies that chloride concentrations correspond to groundwater age (Fig. 9). Correlations between ^{14}C and Cl have also been documented in groundwater from the Eastern View Formation in other regions of the Otway Basin (Love et al., 1994). In assessing this relationship, chloride sources must be considered. That the Cl/Br ratios in the groundwater are similar to those of rainfall preclude significant halite dissolution by the groundwater from the Eastern View Formation, and there are no extensive occurrences of halite in the aquifer matrix.

We propose two explanations of this trend. Firstly, the relationship between $a^{14}C$ and Cl may be explained by mixing of low salinity groundwater that is locally recharged within the valley and high salinity regional groundwater from the Barongarook High. However, the groundwater with high $a^{14}C$ and low Cl also has high ^{3}H activities (0.99–1.47 TU) and if mixing has occurred it must do so at a very slow rate otherwise the resultant groundwater would be expected to contain measurable [3]H. This implies

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that mixing between the shallow groundwater system and the deeper groundwater is limited.

Alternatively the recharge on the Barongarook High may be spatially variable due to the heterogenous nature of the Eastern View Formation or may have undergone changes over time due to climate fluctuations. If evapotranspiration is a dominant process Cl concentrations are likely to be inversely correlated with recharge rates. In the Otway Basin Love et al. (1994) report a decrease in Cl concentrations in groundwater recharged between 18 and 10 ka, followed by an increase in Cl concentrations in groundwater recharged from 10 ka to the present day, which they attribute to increased evapotranspiration rates during a warm Holocene climate. However, in this study decreasing Cl concentrations with increasing a^{14}C would imply that recharge rates on the Barongarook high increased from 10 000 years BP to the present, which is not likely given the warming trend. There is also the possibility that recharge is spatially variable on the Barongarook High, resulting in groundwater flowpaths with a large distribution of groundwater ages and that the high Cl low a^{14}C groundwater is derived from regions with locally low recharge rates (as is the case in the Murray Basin of southeast Australia: Cartwright et al., 2006). Regardless of which model is correct, the chloride measurements provide a useful first order estimate of groundwater residence times.

6 Conclusion

Though widely available water-table measurements offer an insight into recharge, the dynamics of groundwater flow systems and recharge patterns can only be fully understood when combined with geochemical data, in particular radiogenic tracers such as ^{3}H and ^{14}C. These can be used to assess the importance of recharge and discharge in aquifer windows, which in turn defines groundwater pathways and allows the potential fate of pollutants to be assessed. Here shallow (11–42 m) groundwater bores indicate a significant amount of recharge occurs in the Gellibrand River Valley

(90–370 mm yr^{-1}). However, the groundwater at 5–10 m below the water table has ^{14}C ages between 350 and 10 000 years, and below detection ^{3}H activities. Furthermore, there is no indication of water from the river penetrating more than $\sim$ 10 m following flood events. In the Gellibrand River Valley, outcropping aquifer sediments act as a regional discharge zone. Upwards head gradients are maintained for long periods of time and aided by the presence of silts and clays on the floodplain, this limits the depth to which diffuse and localised recharge (via over-bank events and bank exchange) penetrate the aquifer.

There is most likely a shallow local flow system within the Gellibrand River Valley that has limited connectivity with the deeper groundwater. This potentially limits the spread of pollutants such as nitrate and pesticides that may derive from the agricultural activities into the regional groundwater. Future land-use, climate change or groundwater exploitation that occurs on the Barongarook High is likely to affect both the chemistry of groundwater within the valley, and groundwater fluxes to the Gellibrand River, highlighting the importance of regional recharge zones.

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Table 1. Screen depth, Cl, ^{18}O, ^{2}H, ^{13}C, a^{14}C and ^{3}H activities of groundwater samples.

Sample No.	Screen Depth (m)	EC ($\mu S\,cm^{-1}$)	Cl ($mg\,L^{-1}$)	Br ($mg\,L^{-1}$)	Na ($mg\,L^{-1}$)	Ca ($mg\,L^{-1}$)	Mg ($mg\,L^{-1}$)	K ($mg\,L^{-1}$)	HCO_3^- ($mg\,L^{-1}$)	SO_4^{2-} ($mg\,L^{-1}$)	$\delta^{18}O$ (‰ VSMOW)	$\delta^{2}H$ (‰ VSMOW)	$\delta^{13}C$ (‰ PDB)	$a^{14}C$ pMC	1σ	^{3}H TU	1σ
108 899 (a)[a]	29[b]	282	60	0.18	35.1	4.8	2.9	2.2	0.23	0.14	−5.6	−32.7	−21.4	81	0.34	bd[c]	−
108 916 (b)	14.5	197	38.6	0.12	29.3	3.4	4.1	1/9	0.24	0.09	−5.3	−30.4	−22.1	83.3	0.28	bd	−
108 917 (c)	14.5	238	44	0.08	20.3	1.0	2.6	0.7	0.44	0.08	−5.3	−31.1	−21.5	77.8	0.29	bd	−
108 927 (d)	14	430	86	0.07	69.1	16.3	9.9	7.4	0.5	0.36	−5.6	−32	−20	39.5	0.2	bd	−
108 928 (e)	17	446	96	0.08	76.3	19.9	11	8.6	0.58	0.27	−5.5	−33.6	−19.8	40.9	0.21	bd	−
108 933 (f)	11.2	491	121	0.1	84	8.6	5.3	9.1	0.52	0.16	−5.6	−34.1	−20.1	33.8	0.20	bd	−
108 934 (g)	11.5	545	125	0.06	103.8	13.5	8.5	10.5	0.78	0.2	−5.8	−32.4	−20.4	29	0.16	bd	−
108 940 (h)	11.5	243	53	9.02	35.4	3.6	3.21	2.2	0.56	0.11	−5.8	−34	−22.3	64	0.24	bd	−
108 941 (i)	11.5	414	89	0.03	80.3	7.1	3.9	11.5	0.64	0.03	−5.7	−34.3	−21.5	49	0.21	bd	−
110 737 (j)	42	149	31	0.02	16.9	0.9	2.3	0.7	0.08	0.03	−5.11	−29.4	−22.4	100	0.3	1.24	0.06
80 732 (k)	21	200	48	0.1	30	0.33	4.2	0.5	0	0.1	−4.5	−29.7	−24.2	101.5	0.17	1.02	0.03
80 735 (l)	21	217	30	0.03	16.5	0.32	10.5	3.6	0	0.11	−4.2	−29.1	−25.3	100.7	0.17	1.47	0.04
108 935 (m)	11.5	144	27	0.04	19.9	1.7	2.7	0.7	0.12	0.07	−4.8	−31.2	−21.3	88.6	0.17	bd	−

[a] Refers to letter numbers on Fig. 1.
[b] Measured as depth to the middle of the well screen.
[c] ^{3}H activities that are below detection.

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Table 2. Radiocarbon ages of groundwater in the Gellibrand Catchment corrected for calcite dissolution. Uncertainties are calculated varying q by ±0.1 plus the analytical uncertainty of a^{14}C from Table 1.

Sample	q	Radiocarbon Age (years)	Uncertainty
108 899	0.93	1150	+630/−980
108 916	0.96	1190	+360/−940
108 917	0.93	1520	+590/−970
108 927	0.86	6530	+940/−1050
108 928	0.86	6170	+950/−1060
108 933	0.87	7870	+950/−1050
108 934	0.89	9260	+930/−1040
108 940	0.97	3440	+290/−930
108 941	0.93	5310	+630/−980
108 935	0.93	380	+630/−380
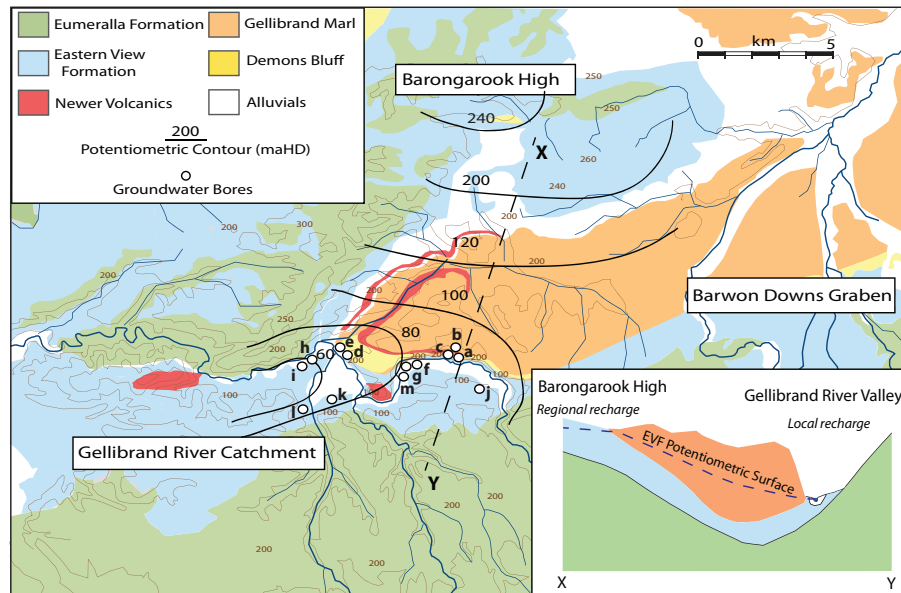
Figure 1. Geology, groundwater flow, and cross sectional view of the Gellibrand River Catchment. Potentiometric contours for the Eastern View Formation are created from groundwater data (Water Resources Data Warehouse, 2013) and are expressed in metres above Australian Height Datum (mAHD). Sampled groundwater bores are also shown. Letters refer to bores listed in Table 1.
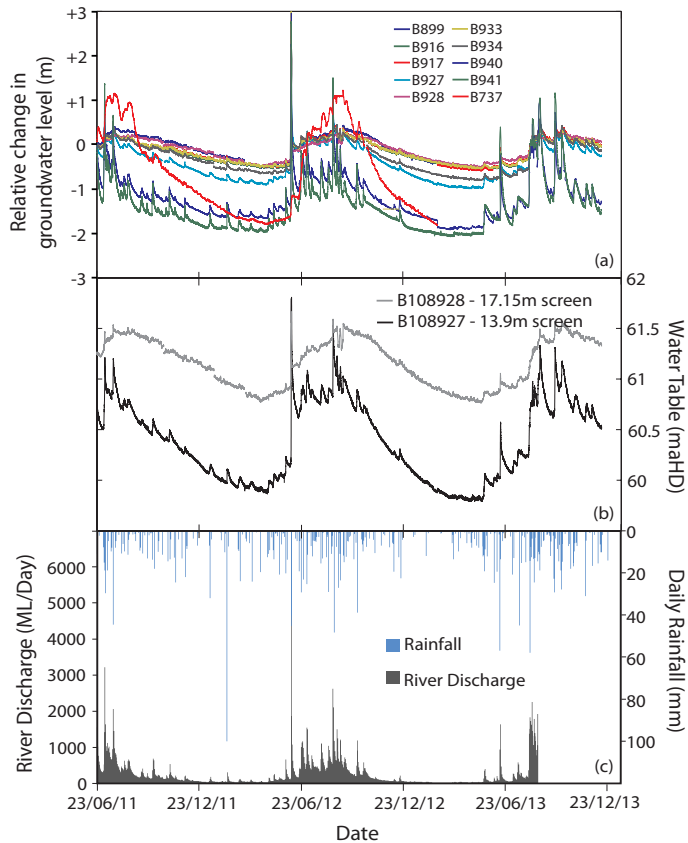
Figure 2. (a) Water table in groundwater bores display clear annual cycles. **(b)** Groundwater head-gradients in the Gellibrand River Valley are upwards implying a discharge zone (Victorian Water Resources Data Warehouse, 2013). **(c)** Flow in the Gellibrand River. Baseflow conditions during summer months transition into high flows in winter following winter rainfall (Bureau of Meterology, 2013).

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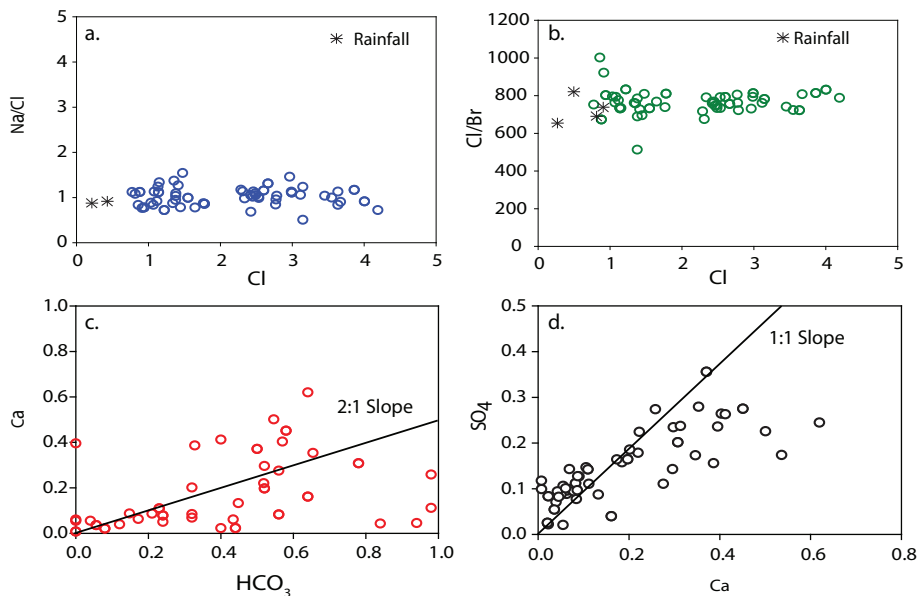
Figure 3. Geochemical characteristics of groundwater in the Eastern View Formation; **(a)** mCl/Br vs. mCl **(b)** mNa/Cl vs. mCl **(c)** mCa vs. mHCO$_3$ **(d)** mSO$_4$ vs. mCa. Rainfall samples are also plotted where measured. Data is from Table 1 with repeat measurements over the sampling period included.
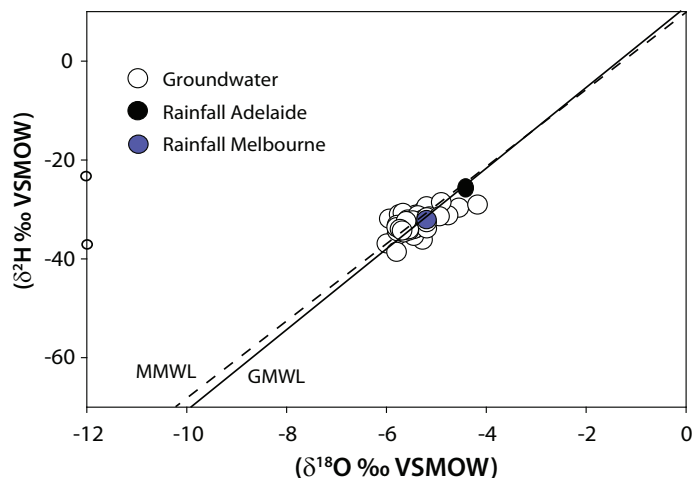
Figure 4. ^{2}H vs. ^{18}O values for the Gellibrand River and surrounding groundwater sampled over March 2011–August 2013 and the weighted average for rainfall from Adelaide and Melbourne. MMWL = Melbourne Meteoric Water Line (Hughes and Crawford, 2012). GMWL = Global Meteoric Water Line. Data is from Table 1 with repeat measurements over the sampling period included.
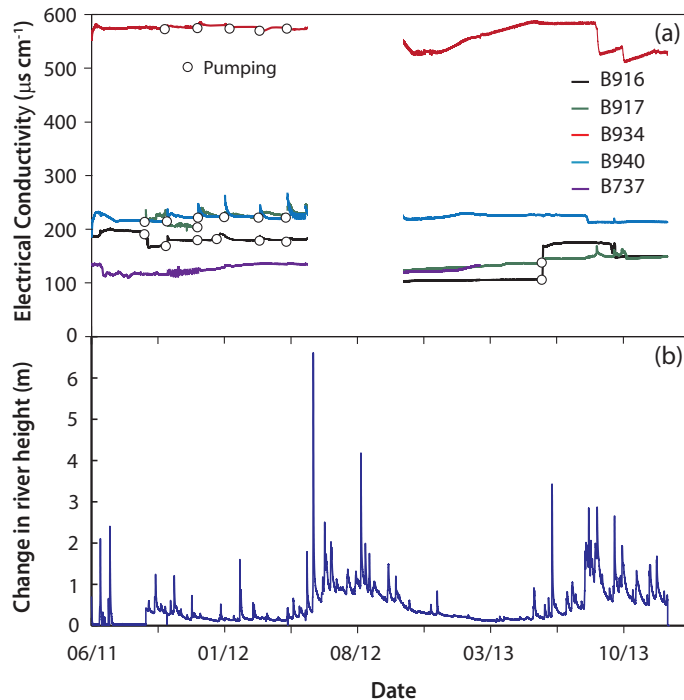
Figure 5. Continuous electrical conductivity monitoring of near-river groundwater and its relationship to **(b)** changes in river height over the study period. Groundwater EC and river level data from deployed Aqua troll 200 (in situ) Data Loggers.
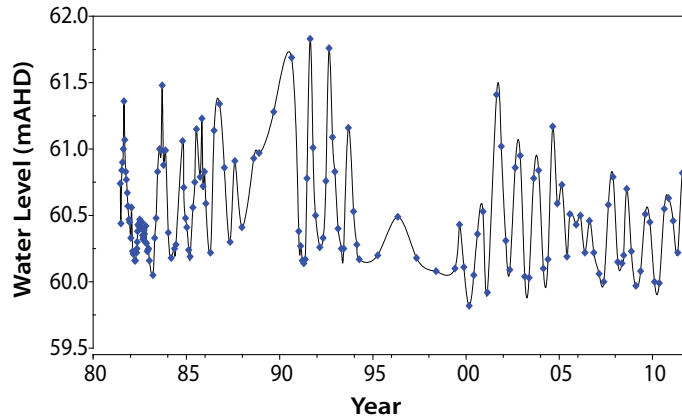
Figure 6. Historical water table fluctuations 1988–2011 for bore 108 927 (Victorian Water Resources Data Warehouse, 2013). The magnitude of annual recharge cycles are coherent with those recorded in data loggers over the study period (2011–2013).
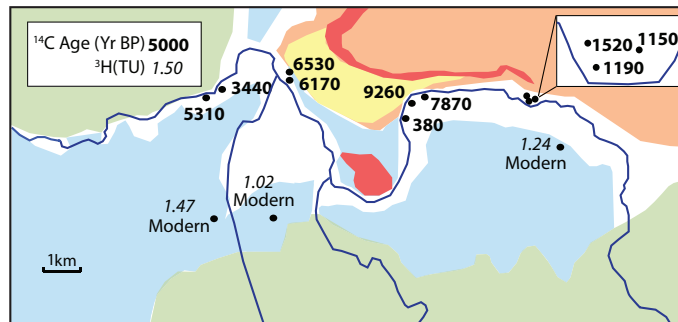
Figure 7. Groundwater residences times within the Gellibrand Valley. Residence times up to 9260 years are found in close proximity to the river. Modern local groundwaters with $a^{14}C >$ 100 pMC are situated back on the floodplain. Data from Tables 1 and 2.

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Using ^{14}C and ^{3}H to understand groundwater flow and recharge

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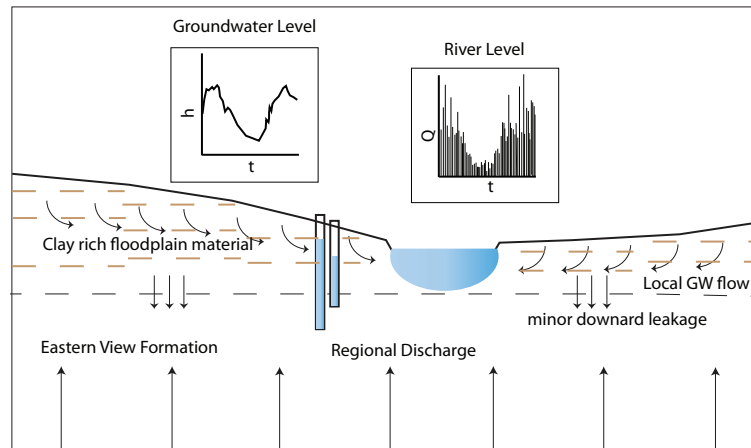
Figure 8. Groundwater flow conceptualisation in the Gellibrand River Valley. Though appreciable amounts of recharge are estimated from bore hydrographs and high river flows, the depth to which recharging waters infiltrate into the Eastern View Formation (downward leakage) is limited by strong upward head gradients, and a floodplain which consists of appreciable amounts of silt and clay.
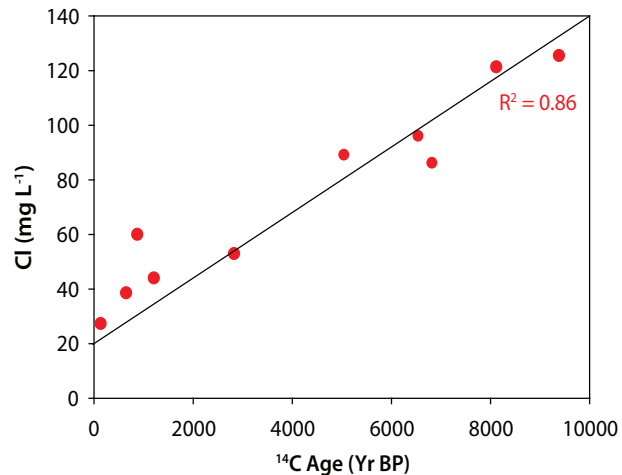
Figure 9. ^{14}C age vs. Cl. ^{14}C ages are taken from the calcite corrected ages in Table 1.