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# Partitioning sources of recharge in environments with groundwater recirculation using carbon-14 and CFC-12

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SUMMARY

Groundwater recirculation occurs when groundwater is pumped from an aquifer onto the land surface, and a portion of that water subsequently infiltrates back to the aquifer. In environments where groundwater is recirculated, differentiation between various sources of recharge (e.g. natural rainfall recharge vs. recirculated water) can be difficult. Groundwater age indicators, in particular transient trace gases, are likely to be more sensitive tracers of recharge than stable isotopes or chloride in this setting. This is because, unlike stable isotopes or chloride, they undergo a process of equilibration with the atmosphere, and historical atmospheric concentrations are known. In this paper, groundwater age indicators ($^{14}C$ and CFC-12) were used as tracers of recharge by surplus mine water that is discharged to streams. Ternary mixing ratios were calculated based on $^{14}C$ and CFC-12 concentrations measured along three transects of piezometers and monitoring wells perpendicular to the creeks, and from dewatering wells. Uncertainty in calculated mixing ratios was estimated using a Monte Carlo approach. Ternary mixing ratios in dewatering wells suggest that recharge by mine water accounted for between 10% and 87% of water currently abstracted by dewatering wells. The calculated mixing ratios suggest that recharge by mine water extends to a distance of more than 550 m from the creeks. These results are supported by seepage flux estimates based on the water and chloride balance along the creeks, which suggest that 85–90% of mine water discharged to the creeks recharges the aquifer and recharge by mine water extends between 110 and 730 m from the creeks. Mixing calculations based on gaseous groundwater age indicators could also be used to partition recharge associated with agricultural irrigation or artificial wetland supplementation.



## 1. Introduction

Groundwater recirculation occurs when groundwater is pumped from an aquifer onto the land surface, and a portion of that water subsequently infiltrates back to the aquifer. Recirculation of groundwater between an aquifer and the land surface can be associated with irrigated agriculture (Brown et al., 2011), artificial wetland supplementation (Searle et al., 2011) or discharge of surplus mine water to streams (Kempe, 1983). In natural environments, water balance estimates are often based on traditional hydrochemical and stable isotopic methods (Harrington et al., 2002; Scanlon et al., 2002; Sharma and Hughes, 1985). In a hydrologic system where water is recirculated between an aquifer and the surface, the application of major ions (e.g. chloride) or stable isotopes as tracers can be limited by the lack of adequate hydrogeochemical variation in potential end-members (Dogramaci et al., 2015; Qin et al., 2011). In such cases, gaseous groundwater age indicators that undergo a process of re-equilibration with the atmosphere through gas exchange may provide a superior alternative to stable isotopes or chloride as tracers of recharge sources.

This paper focusses on mine sites where groundwater recirculation is known to occur. Surplus water produced during mine dewatering (hereafter referred to as mine water) is commonly discharged to natural drainage lines, allowing it to infiltrate into the underlying aquifer. This anthropogenic source of recharge needs to be quantified in order to understand total impact of mining on the aquifer water balance. Our study site is an iron-ore mine in the semi-arid Pilbara region of Western Australia. Prior to mining,

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recharge was dominated by episodic flood events along the creeks (Dogramaci et al., 2012). Discharge of mine water has caused the creek hydrology to shift from ephemeral to perennial flow regimes along the affected reaches (Dogramaci et al., 2015). As a result, natural episodic groundwater recharge through the creeks is now superimposed onto the perennial recharge regime associated with the mine water discharge. Where dewatering occurs between the creek and the mine pit, some fraction of the mine water that recharges to the aquifer through the creeks is potentially intercepted by dewatering wells. This would reduce the net change in the aquifer water balance associated with dewatering operations.

Two groundwater age indicators that could be useful for differentiating sources of recharge in this setting are chlorofluorocarbons (CFCs) and carbon-14 ($^{14}C$). CFCs can be used to date groundwater that recharged between about 1960 and 1990 (Busenberg and Plummer, 1992; Oster et al., 1996; Cook et al., 1995) and have previously been applied as tracers of river water infiltrating into alluvial aquifers (Beyerle et al., 1999) and recharge by irrigation water (Horst et al., 2008; Plummer et al., 2000; Qin et al., 2011). $^{14}C$ has been used extensively as a tracer of groundwater flow and recharge (Hanshaw et al., 1965; Meredith et al., 2012; Pearson and White, 1967; Phillips et al., 1989; Sanford and Buapeng, 1996) and has previously been applied as a tracer of recharge from ephemeral streams in central Australia (Harrington et al., 2002; Shanafield and Cook, 2014). $^{14}C$ activity in streams equilibrates with the atmosphere slower than other gaseous tracers like CFCs because dissolved $CO_2$ is the only fraction of the DIC pool available for exchange with the atmosphere (Bourke et al., 2014b). This slower equilibration, and the different temporal scale of change in atmospheric concentrations, means that $^{14}C$ should provide additional, complementary information, not captured by CFCs.

This paper tests the hypothesis CFC-12 and $^{14}C$ can be used to quantify sources of groundwater recharge at mine sites where groundwater is recirculated. Groundwater samples are partitioned into three principle hydrogeochemical recharge components; old regional groundwater (pre-1960, no CFC-12), recent pre-mining recharge (1960–1996, elevated CFC-12 and modern $^{14}C$) and mine water (post-1996, elevated CFC-12 but old $^{14}C$). Ternary mixing ratios for each sample are calculated from CFC and $^{14}C$ data using a Monte Carlo approach. The lateral extent of recharge by mine water is inferred from mixing ratios in water samples collected from transects of piezometers installed perpendicular to the creeks. These results are compared with independent estimates of the extent of recharge by mine water based on seepage fluxes inferred from the stream water and chloride balance. The relative proportion of mine water re-abstracted by dewatering wells is assessed by calculating ternary mixing ratios in water samples collected from dewatering wells.

## 2. Materials and methods

### 2.1. Site description

The study site is located 80 km north-west of Newman in Western Australia (Fig. 1). The average annual rainfall is around 300 mm $yr^{-1}$ (313 mm $yr^{-1}$ at Newman, 1971–2011, www.bom.gov.au), most of which falls during sporadic high rainfall events that coincide with the passage of cyclones across the coast during summer. Potential evaporation exceeds rainfall, with an average annual potential evaporation of around 3000 mm $yr^{-1}$ (www.bom.gov.au). The two major natural drainage features are Marillana Creek and Weeli Wolli Creek, which flow east and north-east respectively across the Hamersley Basin, draining into the Fortescue Marsh. The target formation for iron ore mining at the study site is a paleochannel deposit known as the Channel Iron Deposit (CID), which in places underlies and is adjacent to the modern creek-lines. Three dewatering well-fields operate across the study site at the locations indicated by sampled dewatering wells shown in Fig. 1. Abstracted water that is surplus to mine requirements is predominantly disposed of by allowing it to infiltrate along the creeks, with some re-injected into the CID aquifer using a series of re-injection wells in the south-east of the study site. There are three discharge outlets within the study site for the disposal of mine water to the creeks; one on Marillana Creek (D1) and two on Weeli Wolli Creek (D2 and D3).

Under natural conditions, groundwater recharge is dominated by cyclonic rainfall events which may only occur every few years. During these events, heavy rainfall leads to rapid runoff and recharge through the high-hydraulic conductivity alluvium in the creek beds, and diffuse rainfall recharge is considered to be negligible (Dogramaci et al., 2012). Stream flow associated with natural rainfall events can last for over a week with stream stage rises of over 2 m not uncommon. In comparison, surface water flow associated with the discharge of mine water extends up to 24 km downstream of the discharge outlets, and produces stream flows of up to 0.8 $m^3 s^{-1}$ with stream stages predominantly less than 0.3 m.

Geologically, the modern drainage lines occur within an alluvial channel that is between 50 and 200 m wide and approximately 5 m thick that is incised into the underlying formations. The creek alluvium is poorly sorted and unconsolidated, consisting of a fine fraction (silt and sand) interspersed within gravel, cobbles and boulders. In places, this alluvial channel is underlain by the CID, which consists of hematitic pisolites in a goethite cement and forms a fractured aquifer. Where not underlain by the CID, the creeks are underlain by a weathered alluvium that consists of moderately sorted silt, sand, clay and minor gravel size fractions. Outside of the creek lines, this weathered alluvium overlies the CID, except where the CID outcrops at the surface. These three formations (channel alluvium, weathered alluvium and CID) are hydraulically connected and are collectively underlain by the Weeli Wolli formation, which consists of Banded Iron Formation (BIF) and dolomite (Kirkpatrick and Dogramaci, 2010). The regional groundwater flow direction is generally from the south-west to the north-east across the study site, sub-parallel to the direction of creek flow. Locally, there is a groundwater mound associated with the creeks, which results in horizontal hydraulic gradients perpendicular to the creeks of 0.02 to 0.03 m $m^{-1}$ measured over distances of up to 500 m from the creeks.

Transects of nested piezometers were installed perpendicular to the creek at three locations (see Fig. 1, Table 1). These locations were chosen to reflect differing hydraulic regimes and lithologies. Transect 1 (JSW) is adjacent to Marillana Creek, which has been receiving on average ~5 GL $yr^{-1}$ of mine water since 1996. The CID aquifer outcrops close to this location and mostly dominates the geology along this transect, with a thin alluvium approximately 2 m thick overlying the CID. A pre-existing nest of two monitoring wells (one screened 0–8 m bgl in the creek alluvium, the other screened 24–72 m bgl in the CID aquifer) was supplemented by the installation of two piezometers approximately 1 km downstream. Piezometer JSW003 was screened 5–6 m bgl within the alluvial aquifer at a distance of 30 m from the creek. Piezometer JSW008 was screened 24–45 m bgl at a distance of 300 m from creek.

Transect 2 (BFS) is adjacent to Weeli Wolli Creek, which has been receiving ~30 GL $yr^{-1}$ of mine water since 2007. This site is at the southern end (down hydraulic gradient) of a re-injection well-field, with the piezometer furthest from the creek within 200 m of the southern-most re-injection well. The weathered alluvium at this site is 15–20 m thick, increasing in thickness away from the creek. This alluvium is underlain by the CID aquifer. Six