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Hydrochemical evolution within a large alluvial groundwater resource overlying a shallow coal seam gas reservoir

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HIGHLIGHTS

- Multivariate statistics and inverse modelling were used to investigate alluvial hydrochemistry
- A focus was placed on the origins of Na-HCO_3, Na-HCO_3–Cl and Na-Cl water types.
- Diffuse recharge and in situ processes are the dominant influences on hydrochemistry
- Alluvial–bedrock interactions were localised and were not major drivers of salinization

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ABSTRACT

A combination of multivariate statistical techniques, simple hydrochemical mixing models and inverse geochemical modelling was used to investigate the major hydrochemical evolutionary pathways of a large alluvial aquifer, the upper Condamine River alluvium, south-east Queensland, Australia. Hydrochemical similarities between alluvium and sedimentary bedrock groundwater imply some mixing between alluvial and sedimentary bedrock aquifers, but spatial assessment showed that this was localised around outcrops of sedimentary bedrock in upstream areas. Within the alluvium, a distinct shift towards a low salinity Na–HCO_3 water type and a brackish Na–HCO_3–Cl water type was obvious in two separate locations. Both of these water types are unique to the alluvium, and inverse modelling shows that they can evolve via a combination of in situ alluvial processes, including diffuse recharge of rainfall or river water or the evolution of basalt-derived groundwater via gypsum dissolution plagioclase weathering, cation exchange and some carbonate precipitation/dissolution. The evolution of these water types is potentially influenced by overlying sodic alkaline soils, and often is associated with a source of sulfate. Evapotranspiration is the dominant salinization process in the alluvium and increases in calcium cations during salinization indicate that brackish Na–HCO_3–Cl groundwater in the underlying Walloon Coal Measures are unlikely to have a major influence on salinization in the alluvium. The most saline water types observed were endemic to shallow zones of the alluvium where evapotranspiration is likely. Results demonstrate that a combination of multivariate statistics and inverse geochemical modelling can be successfully used to delineate hydrochemical pathways in complex hydrogeological settings where a range of environmental and anthropogenic factors may be influencing the evolution of water types with similar hydrochemical compositions.

1. Introduction

Worldwide, alluvial aquifers are renowned as important groundwater resources (Helena et al., 1999; García et al., 2001; Sanchez-Martos et al., 2002; Güler and Thyne, 2004; Lorite-Herrera and Jiménez-Espinosa, 2007; Al-Shaibani, 2008; Chae et al., 2009; Faunt et al., 2009). Characterising the hydrochemical processes within these alluvial aquifers is vital to understanding both the nature and extent of the groundwater resource, as well as its relationships with its surrounding environment, including influences of recharge and anthropogenic activities. Given that alluvial aquifers represent receiving systems, relationships between alluvial aquifers and surrounding aquifers are particularly pertinent in hydrochemical and hydrologic studies. In areas where coal seam gas (CSG), or coal bed methane, resources are being developed, interactions between alluvial aquifers and shallow gas-bearing aquifers is becoming an increasingly important water resource management issue because large volumes of water need to be extracted from the coal seams to release the sorbed gas.

The characteristic Na–HCO_3 or Na–HCO_3–Cl hydrochemical composition, and reduced sulfate concentrations, of CSG groundwater offer a potentially simple means of investigating CSG–aquifer interaction; however, Na–HCO_3 or Na–HCO_3, Cl water types may also evolve via cation exchange, evapotranspiration, interactions with carbonate or via

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processes associated with via with sodic alkaline soils (Rengasamy and Olsson, 1991, 1993; Barbiéro et al., 2002; Van Voast, 2003; Venturelli et al., 2003; Chae et al., 2006; Singh et al., 2006; Hamawand et al., 2013; Owen et al., 2015). While CSG groundwater is often brackish, salinization of groundwater can be a natural process associated with evapotranspiration and hydrochemical evolution, or it may be exacerbated by clearing of native vegetation, irrigation or water extraction (Dixon and Chiswell, 1994; Helena et al., 1999; Zhou et al., 2006; Lorite-Herrera and Jiménez-Espinosa, 2007; Strąpoć et al., 2008; Chae et al., 2009; Choi et al., 2010; Kinnon et al., 2010; Hamawand et al., 2013; Owen et al., 2015). This complexity makes the identification of both CSG–aquifer connectivity indicators and the delineation of hydrochemical evolutionary pathways and water sources problematic, particularly when investigating alluvial aquifers that are often influenced by both in situ processes and interaction with surrounding aquifers.

Multivariate statistical analysis is a useful tool for assessing and explaining hydrochemical variability in complex hydrogeochemical settings, with common techniques allowing hydrochemical water types to be categorised, and the influence of specific parameters on variability to be explained (Kaiser, 1960; Güler et al., 2002; Hair et al., 2006; Cloutier et al., 2008; Templ et al., 2008; Morrison et al., 2011). However, the interpretation of these statistical results can be based on assumptions, and are possibly erroneous, if the statistical outputs are not combined with other analytical techniques and spatial assessments that allow the plausibility of specific processes, such as aquifer connectivity, to be investigated. In some cases inverse geochemical modelling has also been used in combination with multivariate statistics to allow a more descriptive and accurate assessment of hydrochemical processes (Güler and Thyne, 2004; Belkhiri et al., 2010; Carucci et al., 2012). This approach not only allows the characterisation of hydrochemical facies, but also provides a powerful means of delineating major hydrochemical pathways and of describing specific influences on hydrochemical evolution.

In this study, a combination of multivariate statistics and geochemical modelling was used to investigate the hydrochemical evolution within a large alluvial aquifer, the upper Condamine River alluvium, in southeast Queensland, Australia. The alluvium overlies a coal seam gas resource, and underlies sodic alkaline soils. Previous research suggests an influence of surrounding basalt and bedrock aquifer on the alluvial groundwater resource (Huxley, 1982; Hillier, 2012; Dafny and Silburn, 2013). A particular focus of this study was the origins of sodium, chloride and bicarbonate ions and the potential for water types dominated by these ions to evolve via a number of pathways, including via (i) interactions between alluvial groundwater and a range of bedrock aquifers such as coal seam gas groundwater, sedimentary bedrock and basalt aquifers; (ii) diffuse recharge on agricultural soils, e.g., via rainfall, flood flows, or run off; and (iii) in situ hydrogeochemical processes within the alluvium. The origins of sulfate and fluoride ions are also considered.

2. Geological and hydrogeological setting

The Condamine River catchment (here referred to as: the Condamine catchment) is a large surface water subcatchment (30,451 km^2) in the headwaters of the Murray–Darling Basin in southeast Queensland, Australia (Fig. 1). The upper Condamine River alluvium covers an area of around 8500 km^2, and is comprised of a heterogeneous mix of gravels, channels of coarse to fine sands and mixed layer clays, including smectite and kaolinite, as well as carbonate nodules or calcretes, hematite, quartz, feldspars and limonite (Huxley, 1982). The origins of these carbonate deposits have not been studied, but they probably pertain to processes in paleo-palustrine environments, or in shallow environments may be associated with sodic alkaline soils (vertosols and sodosols) that overlie the Condamine alluvium (Rengasamy and Olsson, 1991; Shaw et al., 1994; Biggs et al., 1999; Alonso-Zarza, 2003). The application of gypsum ($CaSO_4 \cdot 2H_2O$) is a common soil management technique in areas where soil sodicity is a problem and this may encourage some carbonate dissolution. Other sources of bicarbonate in alluvial groundwater include the degradation of coal fragments, where the alluvium has incised the bedrock, and in some cases shells (Huxley, 1982).

The alluvium reaches a maximum depth of approximately 130 m, but is generally 30 to 60 m in depth (QWC, 2012). Alluvial aquifers in the headwaters and along the tributaries are generally unconfined, though semi-confined conditions exist where clayey sheetwash overlies the alluvium (Lane, 1979; Huxley, 1982; Kelly and Merrick, 2007; QWC, 2012). An increase in clay content downstream coincides with a decrease in hydraulic conductivity, typically of >30 m/d upstream of Cecil Plains to <30 m/d downstream (Huxley, 1982; Hansen, 1999).

The Condamine alluvium is flanked by a large Neogene alkaline olivine basalt extrusion (Main Range Volcanics) to the northeast and outcrops of the Walloon Coal Measures (WCM) and other Jurassic sedimentary bedrock of the Surat and Clarence–Moreton basins (Fig. 1). Large remnant alluvial deposits overlie the sedimentary Kumbarilla Beds in the west, and colluvial deposits are typical at the base of the basalt extrusions. The sedimentary deposits are comprised of sandstones, siltstones, mudstones and coal, which were deposited in non-marine, fluvial and lacustrine environments (Exon, 1976; Cook and Draper, 2013; Jell et al., 2013). Quartz, kaolinite and mixed layer clays, K-feldspar and plagioclase, carbonates and mixed layers of smectite and illite are typical in most sedimentary deposits (Exon et al., 1972; Grigorescu, 2011). Coal also occurs in most sedimentary deposits, but the most abundant coal seams are found in the WCM (Exon, 1976; Cadman et al., 1998). These sedimentary deposits dip to the southwest beneath the Condamine alluvium, and the WCM and Kumbarilla Beds form the major basement features in some areas (Lane, 1979; Huxley, 1982). In some cases, the alluvium has incised the WCM by up to 130 m within a paleovalley (QWC, 2012).

Early work proposed that stream recharge was the dominant source of alluvial groundwater, although more recent investigations disagree that stream recharge is significant; in any case, recharge is not well understood and diffuse recharge and discharge from surrounding basalt aquifers require further clarification (Lane, 1979; Huxley, 1982; Kelly and Merrick, 2007; KCB, 2011; Barnett and Muller, 2012; Dafny and Silburn, 2013). Dafny and Silburn (2013) provide a comprehensive overview of work to date on the alluvial hydraulics. Since the 1960s, thousands of wells have been drilled into the alluvium and water extraction supports large-scale agriculture in the catchment, primarily for the irrigation of cotton and grain crops. Long-term over-extraction led to a decline in the volume of water present in the aquifer, although the net extraction rate has been significantly reduced in the last decade through improved water resource management policies (Cox et al., 2013; Dafny and Silburn, 2013).

Although the decline in alluvial water levels has produced changes in hydraulic heads that may induce discharge from surrounding bedrock aquifers, alluvial–bedrock interactions have not been confirmed (Huxley, 1982; KCB, 2010; Dafny and Silburn, 2013). In some areas hydraulic heads favour discharge from the WCM to the alluvium, while in others the opposite occurs. Some studies have proposed that the presence of Na–Cl water types and/or increases in salinity in the alluvium are indicators of discharge from the WCM to the alluvium, but the theory remains speculative (Huxley, 1982; Hillier, 2012; Dafny and Silburn, 2013).

Predominantly biogenic coal seam gas (CSG) reserves in the underlying WCM represent a significant economic resource and a number of companies have recently commenced gas production (Draper and Boreham, 2006; Golding et al., 2013; Hamilton et al., 2014). Production of coal seam gas requires large volumes of water to be extracted from the coal seam, in order to reduce hydrostatic pressure and allow the gas to be extracted. Combined with the influence of drawdown due to agricultural development, the extraction of water for CSG production raises some concerns about aquifer connectivity. While the primary gas reservoir that underlies the Condamine catchment is relatively shallow compared to some other areas in the Surat Basin, commercial