Development of an integrated conceptual model of a connected surface water-groundwater system using a hydrochemical approach at Maules Creek, NSW, Australia

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Abstract Hydrochemistry was studied in an alluvial aquifer to develop knowledge of important geochemical processes which could be related to surface water-groundwater interactions in the system. Hydrochemical data consisting of major ions (Na⁺, Ca²⁺, Mg²⁺, K⁺, Cl⁻, HCO₃⁻, SO₄²⁻, H₄SiO₄) and water quality parameters (pH, PCO₂, O₂) were used as natural tracers and to characterise chemical processes in the aquifer system. Primary silicate weathering was found to be a key process, with leaching of salts from the clays also likely to play a role. Using this geochemical understanding and linking it to hydrogeological data and previous investigation of surface water-groundwater interactions, further deductions were made regarding hydrochemical and hydrogeological processes. This enabled development of a stream-reach scale conceptual hydrogeological model which integrates the hydrogeology and hydrochemistry of the aquifer system.

Keywords: hydrochemistry, hydrogeology, chemical weathering, conceptual model, surface water groundwater interactions

1. Introduction

Aquifer heterogeneity often complicates the estimation of water fluxes in sedimentary aquifer systems. This problem is often exacerbated at points of surface water-groundwater exchange (i.e. discharge and recharge processes), and may additionally be seasonally variable (e.g. Sophocleous 2002, Winter et al. 1998). To gain an improved understanding of hydrogeological processes, a hydrochemical approach using naturally occurring chemical species, such as major ions, can be useful. This approach was adopted to develop a process-based conceptual model of aquifer hydrogeology incorporating surface water-groundwater connectivity at Maules Creek, NSW, Australia. This paper focuses on major ion data to develop understanding of important geochemical processes controlling water quality. Subsequently the obtained geochemical knowledge was combined with basic hydrogeological data to deduce important hydrogeological processes and present an integrated conceptual model the aquifer system.

2. Site description

Maules Creek is located in the Namoi Valley, NSW, Australia (Fig. 1), and is situated within an economically-significant irrigated agricultural region. The climate is semi-arid with annual average rainfall approximately 700 mm in the flatter south-west part of the catchment, increasing to 1200 mm towards Mt Kaputar and the Nandewar Ranges in the North. Maules Creek runs from the slopes of Mt Kaputar (1500 m) in the North, across the alluvial catchment and joins the Namoi River in the south-west (Fig. 1). The catchment is underlain by a basement of Permian siltstones, sandstones, conglomerates, coal measures, basaltic and rhyolitic lavas, and tuffs with interbedded shale. These formations outcrop at catchment margins in the north, east and south (DMR 1998). The Nandewar ranges and Mt Kaputar, a 20 million year old Tertiary volcano, are situated in the North of the catchment. Quaternary alluvial sediments varying in thickness from 30 - 100 m unconformably overlie the Permian basement and are comprised of interspersed clay, sand and gravel sequences grading from thick clayey layers further upstream to more frequent sand and gravel layers towards the Namoi River (DNR 1996). In the vicinity of the Namoi River deep coarse gravel and sand deposits comprise the Namoi River paleochannel which is the principal aquifer in the catchment.



Fig. 1. Location of the Maules Creek catchment in New South Wales. Insert shows the location of the investigation transect along Maules Creek which consisted of both surface water and groundwater sampling locations.

The aquifer system can be divided into a lower-yielding clayey up-catchment region (max. 20-30 L/s) and the higher-yielding Namoi River paleochannel (~120 L/s) in the lower catchment (Sinclair et al. 2005). Groundwater flow is broadly towards the Namoi River driven by average horizontal head gradients of approximately 0.003 m/m (Fig. 2, 3). However, the flow pattern is complicated by varying drawdown magnitudes throughout the aquifer due to groundwater abstraction, which is occurring predominantly during spring and summer (October-February). The relatively intensive pumping regimes during these times can lower groundwater levels by up to 11 m, most significantly within the Namoi River paleochannel. These reduced aquifer pressures are often most pronounced in the Lower aquifer zone, and seem to be transmitted vertically through to overlying zones. This has ramifications for surface water groundwater (SW-GW) exchange processes in the vicinity of the Namoi River and in the lower reaches of Maules Creek. Upstream of Elfin Crossing near the top of the transect, groundwater appears to discharge into Maules Creek. Approximately 0.5-7.5 km further downstream of Elfin Crossing surface water is subsequently thought to infiltrate back into the aquifer (Andersen and Acworth 2007a). This seems to be in accordance with decreasing surface water flow in this reach of Maules Creek with the onset of the groundwater pumping season. In most years surface water flow seems to cease altogether with limited flow in the creek bed sediments between seemingly stagnant pools by the end of summer. In the vicinity of the Namoi River where seasonal groundwater pumping is most significant, surface water from the Namoi River appears to infiltrate the streambed during summer months due to aquifer drawdowns (Andersen et al. 2008). However, large gaps remain in the conceptual understanding of hydrogeological and geochemical processes within the aquifer, in the context of surface water-groundwater connectivity.



Fig. 2. Hydraulic head distribution (mAHD) and inferred flow directions in plan in the Upper aquifer (<30 m) in August 2006, also showing distribution of monitoring wells across the catchment.

Fig. 3. Hydraulic head distribution (mAHD) in a cross-section parallel to Maules Creek in August 2006. Also included is the borehole locations, screened depths (red circles) and the basic geology based on drillers logs from monitoring wells installed c.1970-80 (DNR 2006).

3. Methodology

Groundwater samples were taken from a network of monitoring piezometers, 10-100 m deep, (DNR 2006) and from surface waters along an investigation transect along the lower reaches of Maules Creek (Fig. 1, 2) at the beginning of August 2006. Dissolved oxygen (DO) and pH were measured using a HACH portable meter (HQ40d) with a HACH luminescent oxygen probe and pH electrode arranged inline using a flow-through cell. Field analysis for alkalinity was conducted immediately after sample collection on a filtered 25 mL sub-sample using the Gran titration method (Stumm and Morgan 1981) with a HACH Digital Titrator (Model: 16900) and 0.16 N H₂SO₄ cartridges. Major cation samples were collected in 20 mL PE vials and acidified with 2% 5 N HNO₃ and stored at 5°C for preservation, prior to later analysis by ICP-OES using a Perkin Elmer, Optima 3000DV. Anion samples were collected in 10 mL PE vials and frozen for later analysis by IC using a Waters 430 Conductivity detector for chloride. Additional method description on groundwater sample retrieval and collection, and field and laboratory analysis protocols is given in Andersen and Acworth (2007b).

4. Results

4.1 CO₂, pH and dissolved oxygen

The Upper aquifer was characterised by high and relatively constant P_{CO2} (~10^{-1.4}atm) which is likely to be due to respiration processes in the overlying root zone (Appelo and Postma 2005). pH was correspondingly relatively constant ~6.8 (Fig. 4a, b). Deeper samples within the upstream clayey region of the transect, and deep within the Namoi River paleochannel (MW36164) showed lower CO₂ pressures and higher pH than observed for the Upper aquifer, which may reflect the consumption of acidity via weathering processes. However locations near the Namoi River showed uniform pH and P_{CO2} through vertical depth in the aquifer. P_{CO2} observed in surface waters were high upstream of Elfin Crossing (comparable to values in the Upper aquifer) and decreased towards atmospheric equilibrium in a downstream direction, with concurrent increase in pH. This appears to indicate degassing of CO₂charged surface water originating from discharging groundwater upstream of Elfin Crossing. The Upper aquifer was generally oxic within the upstream clayey region of the transect but anoxic close to the Namoi River (Fig. 4c) where higher dissolved organic carbon (DOC) levels produce a steep redox gradient between surface water and groundwater (Andersen and Acworth 2007b). Interestingly, relatively high DO levels (4.5 mg/L) were observed in the Middle aquifer at MW30130 (~60 m depth). Surface water upstream of Elfin Crossing showed DO levels comparable to Upper aquifer groundwater which then increased to atmospheric equilibrium downstream from Elfin Crossing.





Fig. 4. Distribution of a) pH; b) $log(P_{CO2})$; and c) dissolved oxygen (mg/L); along Maules Creek Transect. Distance measured as distance along the transect heading upstream from the most downstream sample location.

4.2 Chloride and alkalinity

The distributions of chloride and alkalinity with depth were found to be relatively similar, showing approximately linear increase in concentration with depth through the Middle and Lower aquifer zones, however more variable and sometimes higher chloride and alkalinity levels were observed in the Upper aquifer (Fig. 5). Along the investigation transect the concentrations of both chloride and alkalinity were variable, and both species showed an elevated region in the Upper aquifer near MW36187 approximately halfway along the transect (Fig. 6). This area also showed elevated nitrate levels (NO₃⁻ ~7.5 mg/L) relative to immediate upstream and downstream regions (where NO₃⁻ ~0 mg/L) as well as concurrent elevations in other species including Na⁺, Ca²⁺, Mg²⁺, Ba⁺, Sr²⁺, and SO₄²⁻. In the Middle aquifer MW30130 showed relatively low chloride and relatively high alkalinity (Fig. 6). In the Lower aquifer, MW36164 showed relatively high chloride and alkalinity (Fig. 6). Other samples within the Namoi River paleochannel showed remarkably uniform chloride and alkalinity signatures through depth and in comparison to surface water (and similar patterns were observed for EC, Na⁺, K⁺, Ca²⁺, Mg²⁺, Sr²⁺).



Fig. 5. Distribution of a) chloride (mM); and b) alkalinity (meq/L) against depth (m below ground surface) through the aquifer in the catchment



Fig. 6. Distribution of a) chloride (mM); and b) alkalinity (meq/L) along Maules Creek Transect. Distance measured as distance along transect from the most downstream sample location.

4.3 Major ions

Na⁺, Ca^{2+} and Mg²⁺ increased approximately linearly with HCO₃⁻ (Fig. 7a, b, c) which seems to indicate mineral weathering processes releasing these species to solution and possibly also leaching of salts from sediments. The Lower aquifer samples from within the Namoi River paleochannel, as well as within suspected gravel beds deep beneath thick clay layers contained relatively low levels of Ca²⁺ and Mg²⁺ compared to the Upper aquifer. K⁺ concentrations were relatively constant with HCO₃⁻ (Fig. 7d). Dissolved silica levels were high and relatively uniform in magnitude (average groundwater concentration 0.64 mM = 18 mg/L), and supersaturated with respect to some silica oxide phases (e.g. SI_{quartz} ~0.9, SI_{chalcedony} ~0.5) (Fig. 7e). SO₄²⁻ showed an approximately linear trend for most samples which plotted relatively lower (Fig. 7f). Ratios of major ions to HCO₃⁻ were also examined, and the (Ca²⁺+Mg²⁺)/HCO₃⁻ ratio (meq/L) showed an approximately linear trend for most Upper aquifer, Middle aquifer and surface water samples, however some of the deeper samples showed relatively lower values (Fig. 7g). The Na⁺/HCO₃⁻ ratio (meq/L) showed an approximately linear trend for most samples (Fig. 7h).

Alkalinity (as HCO₃) can be acquired through various water-rock interactions such as weathering of carbonate or silicate minerals, or by degradation of organic matter (Appelo and Postma 2005). However the aquifer does not seem to contain appreciable amounts of carbonate minerals (except where fluctuating water tables may cause precipitation and re-dissolution of carbonates from solution), and the groundwater is generally sub-saturated with respect to calcite (SI_{calcite} \sim -1.2 to

-0.4). There also does not seem to be appreciable degradation of organic carbon in most of the aquifer away from the Namoi River, indicated by the general presence of DO and NO_3^- and absence of Fe(II) and S²⁻. Therefore it can be assumed that most of the acquired alkalinity is due to primary silicate weathering processes, which are known to be relatively slow and kinetically controlled processes (Appelo and Postma 2005). As a consequence alkalinity should increase with residence time and is therefore used as a progress variable in the plots in Fig. 7.

4.4 PHREEQC model results

A PHREEQC (Parkhurst and Appelo 1999a) batch-reaction model was developed to investigate the influence of primary silicate weathering on the aquifer hydrochemistry. The modelling approach consisted of dissolving increasing amounts of the Ca-plagioclase end-member anorthite into an upstream water composition in a stepwise batch fashion to simulate the geochemical evolution of groundwater along a flowpath subject to the dissolution of anorthite. Anorthite was used as a single idealised primary silicate phase due to the prevalence of Ca²⁺ observed in the groundwater (Fig. 7c), as well as the relatively high weathering rates of anorthite (Appelo and Postma 2005). The geochemical model dissolved 1 mM anorthite into an initial upstream water composition. P_{CO2} was held constant to represent the observed field data of almost constant P_{CO2} in the Upper aquifer (Fig. 6b), which could imply that the dominant weathering reactions occurring in the real system do not substantially consume protons, or that there is an additional source of CO_2 from within the aquifer. Observed Ca² and alkalinity were represented relatively well (Fig. 8a, c). The silica behaviour was also modelled well (Fig. 8b), because Si was conserved in the solid phase by allowing kaolinite precipitation (a secondary weathering product) solubility for when the kaolinite was



exceeded. The modelling of the weathering reaction therefore did not liberate silica to the dissolved phase. pH was also modelled well (Fig. 8d). Based on the observed kaolinite precipitation in the model, this suggests that a weathering reaction such as Equation 1 (Appelo and Postma 2005) could be occurring in the aquifer system.

 $CaAl_2Si_2O_8 + 2H^+ + H_2O \rightarrow Ca^{2+} + Al_2Si_2O_5(OH)_4$



Fig. 8. Modelled evolution of Upper aquifer water compared to observed data for an anorthite weathering model for a) Ca^{2+} ; b) dissolved silica (H₄SiO₄); c) PCO₂; and d) pH.

4.5 Stability diagrams

Stability diagrams were constructed to further investigate the sorts of secondary weathering products that may result from primary silicate weathering. The Ca-silicate stability diagram indicates that a Carich montmorillonite phase may be a stable weathering product for a Ca-plagioclase (Fig. 9a), whereas the Na-silicate stability diagram indicates that kaolinite may be a stable weathering product for a Naplagioclase (Fig. 9b). Based on the volcanic geology and semi-arid climate it may be reasonable to expect formation of montmorillonite (Appelo and Postma 2005) however the complex and heterogeneous geochemistry would mean that many different weathering products would be likely to co-exist. Based on the results of the PHREEQC modelling above combined with the stability diagrams montmorillonite and kaolinite weathering products seem to be likely.



Fig. 9. Stability diagrams for idealised a) Ca-silicate system; and b) Na-silicate system, developed from thermodynamic data sourced from Parkhurst and Appelo (1999b) and Langmuir (1997).

(1)

5. Discussion

5.1 Geochemical processes

Increases in the major cations Na⁺, Ca²⁺ and Mg²⁺ measured against HCO₃⁻, and relatively high dissolved silica levels seem to indicate that primary silicate weathering is a key geochemical control on water composition. Leaching of salts from aguifer sediments is also likely to play a role in providing dissolved material. However, the relative contribution from salt leaching cannot be distinguished. Volcanic rocks such as basalt and rhyolite contain reactive primary silicate minerals which are susceptible to weathering (Appelo and Postma 2005), and both basalt and rhyolite formations are extensive in the up-catchment geology of the region and are likely to have provided much of the Quaternary material that comprises the aquifer. Fragments of primary silicate minerals abound within the sediments along Maules Creek and this sort of detrital material is likely to be ubiquitous within the aquifer matrix. Mineral weathering rates for weathering of relatively reactive primary silicate minerals such as plagioclase feldspars are likely to be comparable to the expected timescales of aquifer residence time. Furthermore, different primary silicate phases may have varying degrees of importance through different aquifer units as a result of varying groundwater residence times. More-reactive calcic plagioclases such as anorthite may play a more important role in the Upper aquifer, whereas in the Middle and Lower aquifer, where aquifer residence times are greater, the influence of less-reactive sodic-plagioclases such as albite may be more significant. This could explain the relatively greater increases in Na^+ compared to Ca^{2+} and Mg^{2+} in some Middle and Lower aquifer samples relative to the Upper aquifer in Fig. 7. Secondary silicate weathering products forming from primary silicate weathering are likely to be kaolinite and montmorillonite. The PHREEQC model suggested that anorthite would weather to kaolinite, with the solution sub-saturated (SI_{Ca-mont} ~ -1.5) with respect to an idealised pure phase Ca-montmorillonite (Parkhurst and Appelo 1999a,b). However stability diagrams, although highly idealised, seem to indicate that the solution may be relatively close to stability with respect to a montmorillonite phase. Overall, there would be a large amount of heterogeneity both in terms of distribution and type of secondary weathering products throughout the aquifer with many different secondary products likely to co-exist. Idealised reactions for weathering of the Ca-plagioclase end member anorthite to montmorillonite, and the Na-plagioclase end member albite to kaolinite are given in Equations 2 and 3 to illustrate the sorts of primary silicate weathering reactions that could be important in the aquifer.

$$7\text{CaAl}_{2}\text{Si}_{2}\text{O}_{8} + 12\text{H}^{+} + 8\text{H}_{4}\text{SiO}_{4} \rightarrow 6\text{Ca}_{0.165}\text{Al}_{2.33}\text{Si}_{3.67}\text{O}_{10}(\text{OH})_{2} + 6\text{Ca}^{2+} + 16\text{H}_{2}\text{O}$$
(2)
anorthite Ca-montmorillonite

$$2NaAlSi_{3}O_{8} + 2H_{2}CO_{3} + 9H_{2}O \rightarrow Al_{2}Si_{2}O_{5}(OH)_{4} + 2Na^{+} + 2HCO_{3}^{-} + 4H_{4}SiO_{4}$$
(3)
albite kaolinite

Primary silicate minerals in the aquifer are also likely to contain K^+ . However, K^+ concentrations were observed to be relatively constant (Fig. 7) and this may be linked to formation of a secondary silicate mineral phase containing potassium acting to control K^+ concentrations in solution. However, the PHREEQC model indicated that the water composition was strongly subsaturated with respect to the K-containing secondary silicate mineral illite (SI_{illite} ~ -3.2). This may indicate that some other secondary silicate mineral phase is locking up K^+ , or alternatively K^+ may be adsorbing to clay minerals (Petrides and Cartwright 2006). The possibility of ion exchange reactions was also considered by examining ion ratios as plotted in Fig. 7g,h. However, no clear trends of concurrent increase and decrease in the concentration of sets of cations could be discerned. Therefore there is little evidence for ion exchange processes providing Na⁺ to solution, and increases in Na⁺ observed in the deeper, down-gradient samples are more likely to be due to weathering of Na-containing primary silicate minerals. The relatively lower levels of Ca²⁺ and Mg²⁺ observed in these deep samples is likely to be related to different geochemical processes across these locations, with some samples located in the Namoi River paleochannel and others within suspected gravel beds beneath thick overlying clay layers.

5.2 Hydrogeology

Various hydrogeological processes can be inferred from the hydrochemical data. The variable and sometimes elevated chloride and alkalinity levels in the Upper aquifer compared to underlying groundwater (Fig. 5) could reflect return flows of irrigation water sourced from deeper aquifer units (Middle and Lower aquifer) recharging the Upper aquifer. Evaporative up-concentration of solutes prior to infiltration is unlikely based on the observed isotope signatures of this groundwater (Andersen et al. 2008). A specific zone of interest in the Upper aquifer was the region of elevated solute load approximately mid-way along the transect in the vicinity of MW36187 (Section 3.2). Solute material could have a geological source as these boreholes are located close to an outcrop of Permian volcanic deposits, however due to the elevated DO and nitrate signatures return flow of irrigation waters may also play a role. In the Middle aquifer at MW30130, high DO (~4 mg/L) and low Cl⁻ could reveal the presence of deep gravel beds connected to hillslopes at the foothills of Mt Kaputar beneath the thick overlying clay layers. Deep in the Namoi River paleochannel at MW36164, relatively high Cl⁻, HCO₃⁻, Ca^{2+} , Mg^{2+} and Na^{+} , $SI_{calcite} \sim 0$ (compared to average $SI_{calcite}$ in the aquifer ~ -0.6), and the detection of sulphide, could indicate that this is geochemically 'old' water from within a sand-gravel aquifer which could be isolated from the main aquifer by the overlying clay layer (Fig. 3). In other samples within the Namoi River paleochannel, similarities in the major ion signature through vertical depth of the aquifer and compared to surface water supports the hypothesis that surface water is a source of recharge to the aquifer, which would be most significant during summer pumping. Furthermore, the hydrochemical data including P_{CO2} , DO, Cl⁻ and HCO₃⁻ have confirmed previous conclusions that groundwater discharge upstream of Elfin Crossing plays an important role in supplying baseflow to Maules Creek.

Fig. 10 summarises a conceptual model of the aquifer system and integrates the hydrochemical and hydrogeological process understanding developed in this investigation, with the surface water-groundwater interactions investigated previously (Andersen and Acworth 2007a,b).



Fig. 10. Conceptual model of hydrogeology and hydrogeochemistry developed from the hydrochemical investigation, and from previous investigations by Andersen and Acworth (2007a,b).

6. Conclusion

This study shows how major ions can be used as natural tracers to characterise geochemical processes occurring in a connected surface water-groundwater alluvial aquifer system in a semi-arid environment. Weathering of primary silicates was found to be a significant process controlling aquifer hydrochemistry, in addition to leaching of salts. However by also considering the role of geochemical processes in relation to hydrogeological processes further insight into aquifer hydrogeology was gained. For example, similarity in the major ion signatures of groundwater compared to surface water in the vicinity of the Namoi River indicates that surface water recharges the aquifer. In addition hydrochemical data suggest that the hillslope regions may potentially contribute to aquifer recharge via deep gravel beds connected to the upper parts of the catchment. The hydrochemical data also indicate that groundwater deep within the paleochannel may be somewhat disconnected from the overlying aquifer. By combining this understanding with previous knowledge of surface water-groundwater interactions at the site, an integrated conceptual model of the aquifer system was developed. Overall this study demonstrates how concurrent hydrochemical and hydrogeological studies using major ion chemistry and basic hydrogeology in this case, can shed significant light on aquifer processes.

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