



# Transient hydrological conditions implied by chloride mass balance in southeast Australian rivers

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## ABSTRACT

A robust correlation between electrical conductivity (EC) values and Cl concentrations in river water from south-east Australia allows detailed Cl fluxes to be calculated from continuous EC and river discharge records. Many Victorian rivers export significantly more Cl than is delivered to their catchments by rainfall. Cl\* is defined as the mass of Cl exported in the rivers relative to that input by rainfall over a multi-year period (Cl\* = 100% indicates that the river exports the same mass of Cl as is input by rainfall). There is a systematic relationship between catchment type and Cl\*. Rivers draining cleared plains have Cl\* values between 50 and 750%, rivers draining volcanic plains have Cl\* values of 770–1600%, whereas rivers with large forested upland catchments have Cl\* values of 50–110%. These values are minima as they do not account for Cl exported by groundwater from the catchments. The calculations are based on long-term (up to 22 year) records that span drought and high rainfall periods. The magnitude of Cl\* is far higher than can be explained by errors in the calculations or variability in rainfall and runoff, and Cl/Br ratios preclude halite dissolution as a source of Cl. The excess Cl reflects hydrological changes in the catchments. Land clearing on the cleared plains has caused the rise of regional water tables which results in the export of Cl from saline groundwater via increased baseflow to the river systems. Drainage systems on the volcanic plains are re-establishing following impoundment by recent (<4.5 Ma) lava flows; Cl which accumulated in shallow groundwater around saline lakes and marshes developed on these volcanic plains is now being exported via the rivers. The upland catchments have undergone less landscape change and may be in chemical balance. The methodology outlined here provides a straightforward assessment of whether catchments are in chemical balance that may in turn indicate whether they are undergoing hydrological changes.

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## 1. Introduction

Assessing if groundwater and surface water systems are in chemical mass balance, that is where the flux of solutes out of a catchment balances solute inputs, is important for understanding whether catchments are changing hydrologically. Solutes in surface water and groundwater are derived from precipitation (rainfall, snowfall, and dry deposition) and the dissolution of minerals. Solutes are exported from catchments via rivers and groundwater, and may also be consumed by mineral precipitation (Fig. 1). Assuming that the boundaries of the surface water and groundwater catchments coincide, the solute balance is:

$$P * X_P + \sum R_X = SW_o * X_{SWo} + GW_o * X_{Gwo} + \Delta ST * X_{ST} \quad (1)$$

(Cartwright et al., 2013), where P is precipitation, SW<sub>o</sub> is surface water outflow, and GW<sub>o</sub> is groundwater outflow (all in m<sup>3</sup>/year). ΔST accounts for changes to the volume of water stored in the catchment

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due to changes in soil moisture, surface storage, or fluctuations in the water table; ΔST > 0 m<sup>3</sup>/year implies an increase in water held in storage due to, for example, a rise in the water table. X is the concentration (kg/m<sup>3</sup>) of the solute of interest in the various components, and the product of the terms gives solute fluxes in kg/year within the catchment. ΣR<sub>X</sub> is the net rate (kg/year) in the catchment for mineral reactions that produce or consume solute X; ΣR<sub>X</sub> = 0 kg/year for solutes that have no mineral sources or sinks. Other terms could be included in the mass balance in specific catchments, for example to take into account groundwater or surface water extraction, diversion of surface water from adjacent catchments, or groundwater inflows from adjacent catchments.

Detailed long-term rainfall records are available for many parts of the world via national meteorological bureaux. River discharges are well constrained in many catchments; for example, the United States Geological Survey Water Data for the Nation programme in the USA ([www.waterdata.usgs.gov](http://www.waterdata.usgs.gov)) and the National River Flow Archive in the UK ([www.ceh.ac.uk/data/nrfa](http://www.ceh.ac.uk/data/nrfa)) contain river discharge records that span several decades or longer. Groundwater levels and river and groundwater chemistry data are also locally available (e.g., United States Geological Survey: [www.waterdata.usgs.gov](http://www.waterdata.usgs.gov) and British Geological

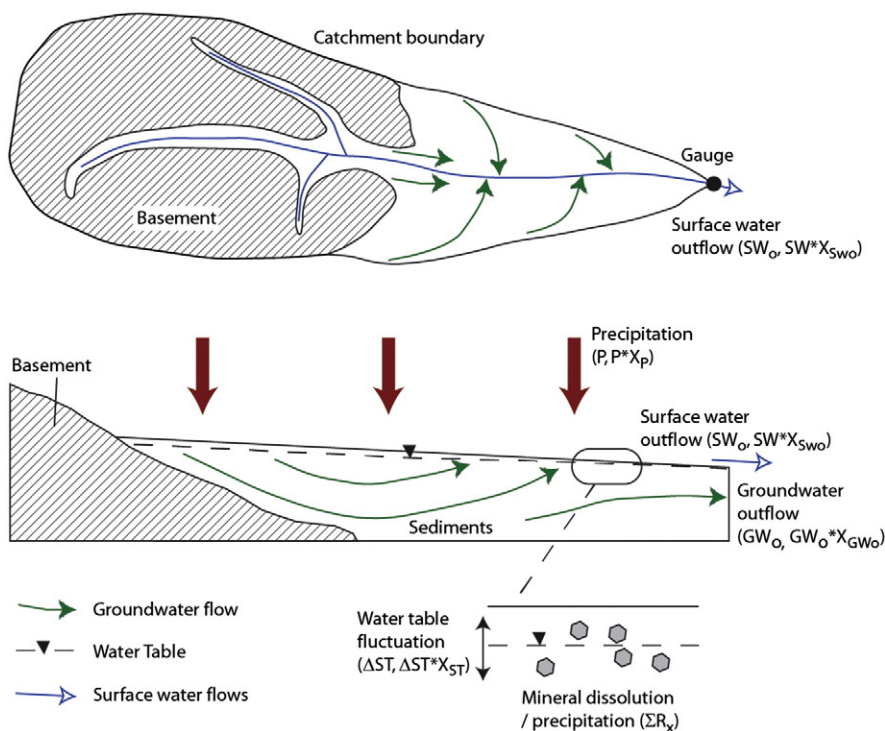


Fig. 1. Schematic depiction of solute and water fluxes in a catchment (modified from Cartwright et al., 2013); terms are defined in the text.

Survey: [www.bgs.ac.uk/research/groundwaterdatabases](http://www.bgs.ac.uk/research/groundwaterdatabases)). Combined with estimates of rainfall chemistry (e.g., Commonwealth Scientific and Industrial Research Organisation, 2012), these data allow many of the solute fluxes in Fig. 1 to be calculated. Groundwater fluxes, however, are less well known (although they may be broadly estimated from hydraulic conductivities and hydraulic heads) and there are very few constraints on catchment-wide rates of mineral dissolution or precipitation. While it is difficult to perform accurate solute balances on catchments without these data, it is possible to identify catchments that export significantly more solutes than are input via rainfall. Although the fluxes are not well known, groundwater generally exports solutes from catchments (i.e.,  $GW_o > 0 \text{ m}^3/\text{year}$ ) and even in closed basins  $GW_o = 0 \text{ m}^3/\text{year}$ . Thus, if the flux of a tracer for which  $\Sigma R_x = 0 \text{ kg}/\text{year}$  out of a catchment via the river system exceeds the input from rainfall (i.e., if  $SW * X_{SW} > P * X_p$ ) the catchment is a net exporter of solutes. There are several reasons that catchments may be net exporters of solutes, including: 1) rising water tables increasing baseflow to rivers that results in the increased export of solutes stored in groundwater via the river systems (Peck and Hurlé, 1973; Allison et al., 1990; Ghassemi et al., 1995); 2) draining of saline marshes which exports solutes that have accumulated over long periods in the soil zone and shallow groundwater (Bennetts et al., 2006; Cartwright et al., 2009, 2013); 3) release of organically-bound solutes during degradation of soils (Bastviken et al., 2006; Svensson et al., 2012) and/or; 4) rivers in coastal areas exporting solutes that accumulated in groundwater from ocean water during previous higher sea levels (Arakel and Ridley, 1986). All these scenarios result from landscapes undergoing hydrological changes where the rivers are exporting solutes that have accumulated in the catchments over decades to millions of years.

### 1.1. Cl mass balance in catchments

The role of biogeochemical processes in soils in retaining or releasing solutes, particularly Cl, is well known (e.g., Bastviken et al., 2006, 2007;

Svensson et al., 2012). Organic matter in soils retains Cl either via ion-exchange or chlorination (Bastviken et al., 2006). If rates of organic matter formation and mineralisation are similar, there may be no net change to soil Cl concentrations. However, where soils are degrading or if soil water residence times or  $O_2$  concentrations change, some of this soil Cl may be released (Oberg and Sanden, 2005; Bastviken et al., 2006; Svensson et al., 2012). Calculation of Cl mass balances due to changes in Cl retention in soils has been mainly carried out in small up-land forested catchments. Assessments of Cl mass balances in larger catchments where groundwater represents a significant store of solutes that are exported through the river systems are less common. Peck and Hurlé (1973) showed that cleared catchments in Western Australia export up to an order of magnitude more Cl than is input from rainfall, whereas adjacent forested catchments were close to being in chemical mass balance. Simpson and Herczeg (1994) determined that the River Murray in Australia exported approximately twice the annual input of Cl from the Murray–Darling Basin, which they attributed to rising water tables and increased export of groundwater-derived solutes via the river system. Cartwright et al. (2013) determined that the Upper Barwon catchment (Victoria, Australia) exported up to 2230% of the Cl input by rainfall, which was attributed to the re-establishment of river systems on a young basalt landscape exporting Cl that had accumulated in shallow groundwater around saline marshes and lakes and/or the rise in the regional water table following land clearing.

In this study we examine multiyear Cl mass balance in 17 catchments with different landuse and geology in southeast Australia. Specifically, we assess whether the catchments are exporting more Cl via the river systems than is delivered annually and are consequently out of chemical and hydraulic equilibrium. Finally, we evaluate the landscape changes that may produce hydraulic disequilibrium, identify regions that contribute most to the surface water salinity, and assess the time-scales over which changes to the hydrologic systems occur. Most of the studies that have addressed solute balances have focussed on anthropogenic changes; here we also examine the impacts of long-term

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