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Roles of forest bioproductivity, transpiration and fire in a nine-year record of cave dripwater chemistry from southwest Australia

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Abstract

Forest biomass has the potential to significantly impact the chemistry and volume of diffuse recharge to cave dripwater via the processes of nutrient uptake, transpiration and forest fire. Yet to-date, this role has been under-appreciated in the interpretation of speleothem trace element records from forested catchments. In this study, the impact of vegetation is examined and quantified in a long-term monitoring program from Golgotha Cave, SW Australia. The contribution of salts from rain and dry-deposition of aerosols and dissolved elements from soil mineral and bedrock dissolution to dripwater chemistry are also examined. This study is an essential pre-requisite for the future interpretation of trace element data from SW Australian stalagmite records, whose record of past environmental change will include alterations in these biogeochemical fluxes. Solute concentrations in dripwater vary spatially, supporting the existence of distinct flow paths governed by varying amounts of transpiration as well as nutrient uptake by deeply-rooted biomass. Applying principal components analysis, we identify a common pattern of variation in dripwater Cl, Mg, K, Ca, Sr and Si, interpreted as reflecting increasing transpiration, due to forest growth. Mass-balance calculations show that increasing elemental sequestration into biomass has the largest impact on SO_4, providing an explanation for the overall falling dripwater SO_4 concentrations through time, in contrast to the transpiration-driven rising trend dominating other ions. The long-term rise in transpiration and nutrient uptake driven by increased forest bioproductivity and its impact on our dripwater chemistry is attributed to (i) the post-fire recovery of the forest understorey after fire impacted the site in 2006 CE; (ii) and/or increased water and nutrient demand as trees in the overlying forest mature. The impact of climate-driven changes on the water balance is also examined. Finally, the implications for interpreting SW Australian speleothem trace element records are discussed.
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Keywords: Golgotha Cave; Fire on karst; Speleothem; Forest bioproductivity; Cave dripwater chemistry; Mass balance

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1. INTRODUCTION

Speleothem trace element records serve as proxies for diverse information including recharge (McDonald et al., 2004; McMillan et al., 2005; Johnson et al., 2006; Tremaine and Froelich, 2013), soil and bedrock weathering (Musgrove and Banner, 2004; Hori et al., 2014; Rutlidge et al., 2014), aerosol flux (Frisia et al., 2005), organic matter composition and flux (Hartland et al., 2012) and cave PCO_2 (Wynn et al., 2014a; Treble et al., 2015). These records are readily obtained at high resolution (seasonal or better) using laser ablation, secondary ionisation mass spectrometry (SIMS) or synchrotron instruments (Fairchild and Treble, 2009). Such data may reveal annual chemical laminae which are useful for chronological purposes (Treble et al., 2003; Sundqvist et al., 2013; Orland et al., 2014) or to identify seasonal recharge characteristics (Borsato et al., 2007; Fairchild et al., 2010).

Trace elements are thus an excellent complement to stable oxygen and carbon, given their potential to reduce the uncertainties of isotopic records when interpreted in multi-proxy fashion (Fairchild and Treble, 2009). For example, Cruz et al. (2007) and Treble et al. (2003) both draw on trace element data to distinguish meteoric source versus trajectory effects in stalagmite $\delta^{18}O$ records. Speleothem trace element records are also used as paleoenvironmental records in their own right, particularly in situations where the interpretation of stable isotopes might be complicated by mineralogical changes (e.g. McMillan et al., 2005).

Ions in cave dripwater may be derived from incoming meteoric precipitation, dust, sea spray and soil/bedrock leaching (Baker et al., 2000; Fairchild and Treble, 2009; Hartland et al., 2012), although their ultimate concentrations may be dominated by subsequent water/rock interactions occurring along individual water flow paths associated with the hydrological routing (Tooth and Fairchild, 2003; Baldini et al., 2006; McDonald and Drysdale, 2007). Their appeal as proxies lies in their sensitivity to various hydrological and biogeochemical processes, which have been validated via modern speleothem studies. For example, Treble et al. (2003) demonstrated that Mg concentrations in a SW Australian stalagmite clearly responded to a significant decrease in rainfall. In this case, the mechanism of prior calcite precipitation (PCP) following reduced infiltration was invoked. PCP describes the precipitation of calcite from a solution before it reaches the stalagmite as the fluid equilibrates with lower PCO_2 along its flow path in the unsaturated zone (Fairchild et al., 2000). Removal of Ca causes fluid (and hence stalagmite) ion/Ca ratios to rise.

Studies of *vegetation-related* processes have centred on influences of soil PCO_2 or retention of trace elements in the ecosystem-soil environment. Qualitatively, an increase in speleothem $\delta^{13}C$ is commonly attributed to a lowered soil PCO_2 caused by loss of vegetation cover (Fairchild and Frisia, 2014). Seasonal loss of colloid-associated trace elements from soils can be associated with a peak in fluorescent organic matter (Baker et al., 2008) and a multi-year record of these effects, at the forested Alpine site of Grotta di Ernesto, was linked to an historic episode of deforestation (Borsato et al., 2007). Reforestation above a modern mine cavity site in the UK during the 20th century led to a reversal of both $\delta^{13}C$ and trace element signals (Baldini et al., 2005; Fairchild and Hartland, 2010). Progress in quantifying solute storage effects has been made at the Ernesto site where the late 20th century rise in sulphur pollution is lagged and attenuated in dripwater and speleothem records by around 20 years, due to storage in soil and vegetation (Wynn et al., 2013; Borsato et al., 2015). This work was combined with studies of S distribution in wood (Fairchild et al., 2009; Wynn et al., 2014b) to study environmental S loading using trees and speleothems in parallel.

In water-stressed environments such as Western Australia, the potential for transpiration to raise solute concentrations in the vadose zone, hence in dripwater, is expected to be high. This region therefore offers an opportunity to quantify, for the first time, the effects of transpiration as well as uptake of ions as nutrients by vegetation and hence determine the impact of vegetation dynamics on dripwater and speleothem chemistry. Although climatically imposed changes in water balance can be expected to have an impact, there is also the potential that a system in steady-state will be disrupted by fire or by land-use change with implications for dripwater chemistry.

In this paper, more than nine years of cave monitoring data are presented, focusing on chemistry data from five drip sites in the long-term monitoring study at Golgotha Cave, SW Western Australia. The site is situated in a forested catchment and is a well-characterised system (Treble et al., 2013; Treble et al., 2015) with a relatively simple hydrogeology compared to other karst environments providing clearer distinction of hydrogeochemical processes. For example, studies in karst systems where fracture-dominated flow occurs in mature limestones is typically associated with more complex flow dynamics (Baker and Brunsdon, 2003). Principal components analysis (PCA) is used to highlight the main processes influencing dripwater solute concentrations and via a quantitative mass-balance approach the main sources of solutes and changes in their input to the dripwaters are identified. The impact of the deeply-rooted vegetation on solutes in the vadose zone e.g. via transpiration and nutrient uptake is demonstrated. The relative impact of post-fire vegetation re-growth, as well as climate-driven changes in the hydrologic water balance, on our dripwater chemistry, are also examined.

2. SITE DESCRIPTION

2.1. Golgotha Cave location and climate

Golgotha Cave (36.10°S 115.05°E; Fig. 1A) is developed in the Spearwood System of the Tamala Limestone, a coastal belt of dune limestone that extends along >1000 km of the SW Western Australia coastline (Fig. 1B). The Spearwood System comprises medium to coarse grained Quaternary aeolian calcarenite mantled by weathered siliceous dune sands (Hall and Marnham, 2002). Golgotha Cave lies in the winter rainfall zone of southern

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