



Modelling karst vadose zone hydrology and its relevance for paleoclimate reconstruction



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ABSTRACT

Understanding past climatic changes allows us to better understand how our planet will evolve in the future. One important source of information on paleoclimate is the analysis of speleothems that develop in karst caves and conduits due to the dissolution and precipitation of calcite. However, there are many uncertainties in paleoclimatic reconstruction with speleothems; one of them being hydrological variability. Up to now only few studies have considered the impact of hydrological variability on speleothem formation and composition. This review paper will provide an introduction to hydrological processes that have the potential to affect speleothem composition and the hydrological modelling approaches that are able to account for them. It presents the current state of knowledge on paleoclimatic reconstruction using speleothems and shows that many important flow and transport processes have not yet been included in the interpretation of these archives, mostly due to a lack of field information to parametrize them. Possible directions of future research efforts therefore include a better exploration of karst vadose zone processes and new approaches to incorporate this information into simulation models. Finally, we foresee the exciting advances in reconstructing paleohydrology using karst hydrology models combined with speleothem growth rate and geochemical composition to understand how past climate changes affected the hydrological cycle and water availability.

1. Introduction

Karst systems are characterised by strong feedbacks of climate, hydrology, biology and geology (Ford and Williams, 2013; Goldscheider, 2012). They develop due to chemical weathering of carbonate rock that is driven by dissolved atmospheric and biogenic carbon dioxide, a process referred to as karstification (Király, 2003). Karstification widens fissures and cracks that evolved by tectonic processes and physical weathering. It results in a strong heterogeneity of hydraulic properties (Bakalowicz, 2005; Worthington et al., 2016) that strongly affect the surface and subsurface water flow and storage dynamics (Goldscheider and Drew, 2007), as well as the reactive transport and water-rock interactions (Kaufmann and Braun, 2000). The resulting complexity has been providing a challenge for karst research for decades (Hartmann, 2016; Hartmann et al., 2014a; Király and Morel, 1976; Kovacs and Sauter, 2007; Sauter et al., 2006; Teutsch and Sauter, 1991; White, 1977). However, preferential infiltration processes and the high storage capacities that evolve by karstification also provide favourable conditions for drinking water development from karst aquifers (Andreo et al., 2006) and in some countries, karst aquifers provide up to 50% of

the total drinking water (COST, 1995).

Another characteristic is the precipitation of calcium carbonate in caves in the vadose zone – speleothems (Hill and Forti, 1997). Uniquely, these deposits can be preserved from under eroding landscapes for millions of years (Meyer et al., 2009; Sniderman et al., 2016). And they entrap radioactive U, Th and Pb isotopes in a closed system, allowing them to be precisely dated over the whole of Earth history (Cheng et al., 2016; Woodhead et al., 2010). At the same time, the speleothems contain geochemical tracers which contain clues of the environment at their time of deposition (Fairchild and Baker, 2012). Biological or sedimentary archives may contain geochemical or physical evidence of past environments. Speleothems are one such archive, and their record of past environments can be used as proxies for past climate change. Proxy paleoclimate records can be used to understand the magnitude and frequency of past changes (Cheng et al., 2016), to provide independent comparison to climate model simulations (Goosse et al., 2012), and to quantify forcing mechanisms (Stap et al., 2016). Speleothems are precisely datable, can contain annual lamination, and can be analysed at sub-annual resolution (Baker et al., 1993b; Broecker et al., 1960; Orland et al., 2014). Understanding how the geochemical

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signal contained within them relates to climate requires an understanding of karst unsaturated-zone hydrology (Kaufmann, 2003).

Speleothems, such as stalactites and stalagmites, are formed from karst waters which are supersaturated with calcium carbonate (Holland et al., 1964). This supersaturation derives from the dissolution of limestone from waters which have reacted with carbon dioxide, which originates from the atmosphere (approximately 400 ppm) but is also typically present at higher concentrations in both the soil and vadose zone due to microbial and root respiration (Liu et al., 2007; Matthey et al., 2016). Speleothem growth mechanisms not requiring soil and vadose zone carbon dioxide are possible: for example, carbonate dissolution due to sulphide oxidation can permit speleothem growth in glaciated regions (Atkinson, 1983; Häuselmann et al., 2015; Spötl and Mangini, 2007). These mechanisms are considered uncommon, but in any case, still require a karstic water supply. Therefore, periods of speleothem growth can be used as the most basic paleoclimate reconstruction tool, as they provide evidence of water recharge to the karst (Baker et al., 1993a; Jo et al., 2014). However, paleoclimatic information can also be contained within the speleothem calcium carbonate where stable isotopes of carbon and oxygen are the most widely used proxies for paleoclimate reconstruction (Cheng et al., 2016). Impurities within the speleothem may also be paleoclimate proxies, for example trace element concentrations and isotope ratios, flood layers, and organic biomarkers (e.g., Blyth and Schouten, 2013; Denniston et al., 2015). The rate of speleothem growth can also be used in paleoclimate reconstruction (Baker et al., 2015). Data archived in the World Data Center for Paleoclimatology (<https://www.ncdc.noaa.gov/data-access/paleoclimatology-data/datasets/speleothem>) comprises oxygen isotope data (~85% of all data archived), followed by annual growth rate data (~10% of all data archived).

Speleothem oxygen isotopic composition is a function of numerous processes. Oxygen isotope variability initially follows that of isotopic composition of rainfall, and specifically the rainfall recharged to the vadose zone. Predominantly, this is from diffuse recharge from rainfall or snowmelt. This recharge water subsequently undergoes mixing in the soil, epikarst and vadose zone, and the oxygen isotopes can be fractionated by processes occurring in the soil, vadose zone and cave (Cuthbert et al., 2014; Scholz et al., 2009). A final temperature-dependent fractionation occurs during incorporation into the speleothem calcite (Coplen, 2007; Dietzel et al., 2009; Kim and O'Neil, 1997). As a result, although speleothem oxygen isotopes clearly record global climate change over glacial transitions (Cheng et al., 2016), they are a mixed proxy of temperature and recharge processes, and their interpretation can be ambiguous. For reviews of speleothem oxygen isotopes we refer the reader to McDermott (2004) and Lachniet (2009), and of speleothem paleoclimate proxies in general to Fairchild et al. (2006) and Baker and Fairchild (2012). Hydrological modelling of oxygen isotope systematics provides a potential framework for interpreting and quantifying speleothem oxygen isotope records (Bradley et al., 2010). A hydrological modelling framework for the annual growth rate as a paleoclimate proxy would also assist in the interpretation of this mixed proxy of climatic and karst hydrological processes. However, there is presently little overlap between speleothem and paleoclimate research and karst vadose zone research and adequate approaches to develop and test such frameworks are still missing.

This review paper will bring together experiences from modelling and paleoclimate reconstructions using speleothems in order to provide a base for future advances in paleoclimate as well as paleohydrology reconstructions. We will firstly review the current state of knowledge of karst hydrology and solute transport modelling with a particular focus on the hydrology between the land surface and the cave, i.e. the vadose zone. Secondly, we will elaborate the current state of knowledge of paleoclimatic reconstruction by speleothems, which is finally followed by a list of current research gaps and directions to better use hydrological understanding and modelling to advance paleoclimate and paleohydrology reconstructions.

2. Karst system hydrology and modelling

2.1. Karst processes

Most karst develops from the CO₂-driven dissolution of carbonate rock (limestone, dolostone). Consequently karst surfaces and subsurface are characterised by a strong water-rock interaction when longer time scales are considered (Hartmann et al., 2014a). In the case of calcite (CaCO₃), carbonate rock dissolution can be expressed by



This is resulting in calcium (Ca²⁺) and bicarbonate (HCO₃⁻). Other factors such as mineralogical or chemical purity of the rock, CO₂ partial pressure and temperature also influence calcite dissolution (Buhmann and Dreybrodt, 1985; Goldscheider and Drew, 2007).

Due to carbonate rock dissolution, two types of porosity can be distinguished: on one hand a matrix porosity comprised of intergranular porosity and small fissures, and on the other hand the conduit porosity of the dissolution-enlarged fissures and faults (Ford and Williams, 2013). They result in the heterogeneous hydrological flow and storage behaviour of karst systems (Bakalowicz, 2005) expressed by sinking streams (Goldscheider and Drew, 2007), storage and flow concentration on the epikarst (Aquilina et al., 2006; Williams, 1983), overflow springs (Barberá and Andreo, 2011; Worthington, 1991), concentrated and diffuse groundwater flow and exchange in the fissure matrix and the karst conduits (White, 2003), and many more particular processes as described in several books and review papers (Ford and Williams, 2013; Goldscheider and Drew, 2007; Hartmann et al., 2014a).

2.2. Simulation of karst hydrology

Presently, models to simulate karst hydrology consider a wide range of the processes that result from karstification. For instance, they consider sinking streams (Doerfliger et al., 2008; Le Moine et al., 2008) or epikarst processes (Hartmann et al., 2012; Kiraly et al., 1995; Tritz et al., 2011). Overflow springs were included into hydrological models by Charlier et al. (2012) and Chen and Goldscheider (2014), while Butscher and Huggenberger (2008) or Reimann et al. (2011) include the exchange of groundwater between matrix and conduits. Most of these approaches are attributed to the group of lumped or distributed approaches (or somewhere in between), referring to the spatial discretization in the model. Distributed models discretize the karst system to rectangular or triangular grid cells and calculate groundwater level and flow for each of them, while lumped approaches consider the entire karst system by a set of reservoirs, each of them representing a sub-system (e.g., epikarst, fissure matrix or conduits) in a more conceptual way. Consequently, lumped models rather provide average or “effective” information on the flow and storage behaviour of the system. There are many reviews of the different karst modelling approaches including more detailed and specific information (Ford and Williams, 2013; Ghasemizadeh et al., 2012; Hartmann et al., 2014a; Kovacs and Sauter, 2007; Sauter et al., 2006; White, 2007).

2.3. Simulation of solute transport and hydrochemical processes

Modelling of solute transport and hydrochemical processes has been used for various purposes (Fig. 1). Water quality predictions (Charlier et al., 2012; Hartmann et al., 2016), interpretation of hydrochemical dynamics or tracer tests (Birk et al., 2005, 2006; Long and Putnam, 2009), multi-variate model evaluation and calibration (Hartmann et al., 2013b; Oehlmann et al., 2014), transit time estimation (Einsiedl, 2005; Long and Putnam, 2004; Maloszewski et al., 2002), speleogenesis simulations (Bauer et al., 2003; Hubinger and Birk, 2011; Liedl et al., 2003), and paleoclimate reconstruction (Bradley et al., 2010). In the case of distributed modelling, the advection-dispersion equation can be

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