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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 (Kiraly, 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; Kiraly 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; Mattey 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 CO_2-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 ($CaCO_3$), carbonate rock dissolution can be expressed by

$$CaCO_3 + H_2O + CO_2 \leftrightarrow Ca^{2+} + 2HCO_3^{2-} \quad (1)$$

This is resulting in calcium (Ca^{2+}) and bicarbonate (HCO_3^-). Other factors such as mineralogical or chemical purity of the rock, CO_2 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 serval 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 subsystem (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

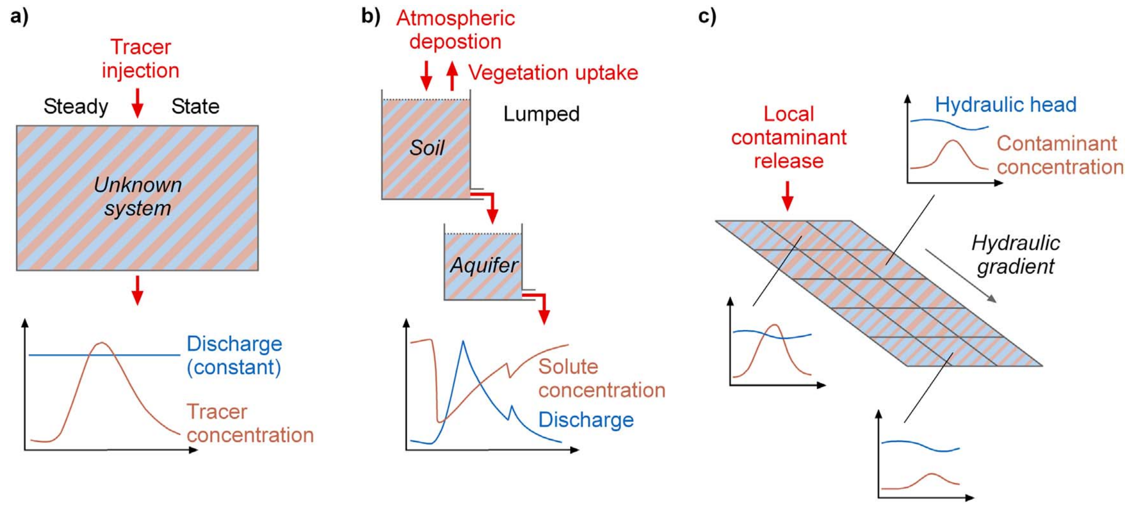

Fig. 1. Elaboration of the 3 solute transport simulation approaches; (a) lumped parameter approach to simulate an artificial tracer concentration assuming stationary hydrological conditions, (b) solute mass balance within a lumped reservoir model to simulate hydrological and hydrochemical dynamics with a constant atmospheric solute input, and (3) distributed modelling of a point input of a contaminant and its adjective and dispersive transport along the hydraulic gradient.

applied (here represented for one dimension):

$$\frac{\partial c}{\partial t} = -v\frac{\partial c}{\partial z} + D\frac{\partial^2 c}{\partial z^2} \tag{2}$$

With c as the solute concentration, v the flow velocity, D the dispersion coefficient, z the location, and t the time. Depending on the degree of karstification, v and D will vary across the karst system, resulting in a complex solute transport behaviour. Modifications of Eq. (2) also allow for including exchange between matrix and fissures (Ghasemizadeh et al., 2012; Maloszewski, 1994) and rock dissolution processes (Bauer et al., 2003). The most common way of integrating solute transport into lumped models is the application of solute mass balance on each of their reservoirs assuming complete and instantaneous mixing. Hence, advection and dispersion are not considered explicitly as in Eq. (2) but implicitly by the sequential mixing along the flow path given by the structure of different reservoirs of a chosen lumped model. As with distributed hydrochemical simulations, exchange between matrix and conduits, as well as rock dissolution, can be considered (Butscher and Huggenberger, 2008; Hartmann et al., 2013a). A third group of solute transport modelling approaches are steady state approaches that assume stationary flow conditions to estimate the shape parameters of travel time distribution functions. Usually applied with water isotopes (^{2}H, ^{3}H, ^{18}O) or artificial tracer concentrations, these so-called lumped parameter approaches allow estimating basic characteristics of the solute transport behaviour of a karst system and its average hydraulic properties (Maloszewski et al., 2002).

3. Soil, vadose zone and cave processes

3.1. Hydrology of the vadose zone

Up to now karst studies most often considered entire karst systems, as they were focusing on water resources and water quality assessment. More attention was given on the unsaturated zone processes when groundwater recharge estimations (Andreo et al., 2008; Ireson and Butler, 2013) or cave water dynamics (Markowska et al., 2015; Sheffer et al., 2011) were investigated. Most of them rely on a common understanding of the functioning of soil, epikarst, and unsaturated zone. The soil has an important hydrological role, being the first potential store of precipitation. The soil storage capacity depends on its physical properties, such as soil depth and soil clay and organic matter content. The volume of water that can be stored in the soil at a particular time depends on the storage capacity and the soil moisture content, the latter being determined by antecedent conditions. Due to subsurface heterogeneity, soil thicknesses show high variability, as well as their hydraulic conductivity (Ries et al., 2015), resulting in spatially variable soil storage capacities and an interplay of diffuse and preferential flow processes (Hartmann, 2016).

The epikarst below is an area of higher carbonate rock dissolution activity due to enriched CO_2 concentration in the infiltrating waters (see Subsection 3.2). It is considered as a temporary storage and distribution system for infiltrating water on its way towards groundwater recharge (Smart and Friederich, 1987; Williams, 1983). Perched water tables can occur that concentrate infiltrating waters towards enlarged fissures and cracks (Mangin, 1975; Perrin et al., 2003). Similarly, the unsaturated zone can be seen as a system of coupled flow systems, one channelling the concentrated flow in enlarged fissures and cracks, the other, less permeable with diffuse matrix flow in the fissures (Kordilla et al., 2012). In both the soil and unsaturated zone, the role of vegetation should be remembered, and in particular the effect of trees and their root networks. Depending on the species, trees may use soil, shallow or deep vadose zone water, in some instances affecting drip water discharge to 30–50 m depth (Coleborn et al., 2016). The primary effect of trees on the karst system is therefore to change the water balance through loss of water by transpiration (Fig. 2).

The heterogeneous flow and storage behaviour in the unsaturated zone results in different drip rate flow characteristics (Bradley et al., 2010): There are drips that are fed (1) solely by the fissure matrix, (2) by the matrix and enlarged fissures or cracks, (3) by water that was routed through a reservoir while traveling through matrix or conduits, (4) or by water that passed through an "overflow" water storage, such as the soil or depressions in the fissures, that has to fill before water reaches the drip. Similarly, Smart and Friederich (1987), Arbel et al. (2008) and Lange et al. (2010) defined different types of drips by their dynamical behaviour: (1) post-storm drips that only activate after considerable rainfall events, (2) seasonal drips that are only active during the wet season of the hydrological year, (3) perennial drips that are active all year, and (4) overflow that only activate after infiltrated volumes exceed a certain threshold.

3.2. Calcite dissolution and precipitation

The soil zone typically contains high concentrations of CO_2 gas that has been produced by root and microbial respiration. Soil CO_2 production is a function of temperature and soil moisture, and soil CO_2

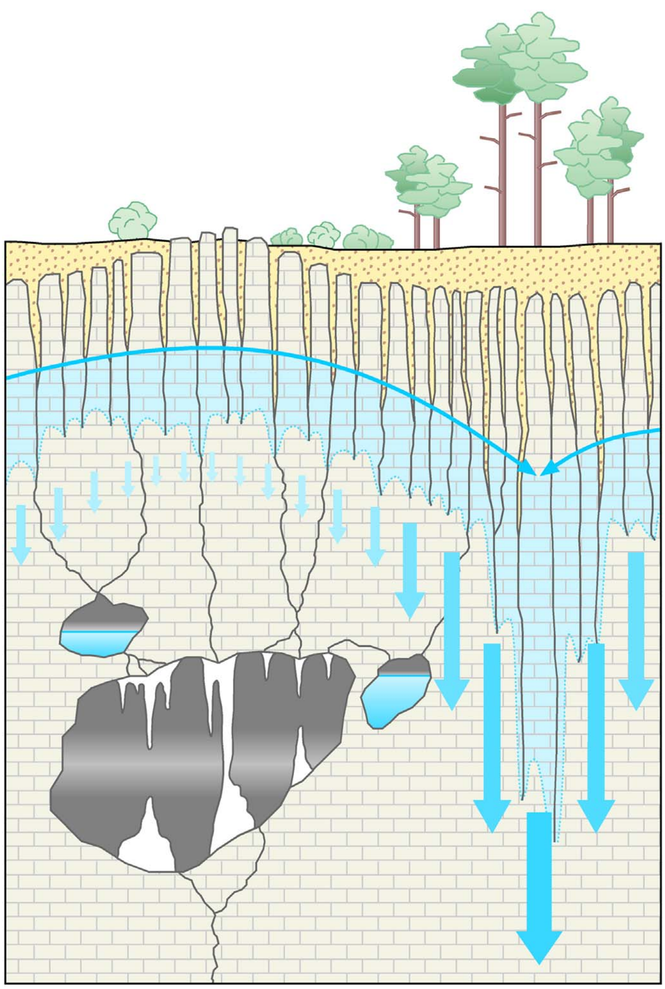

Fig. 2. Schematic description of karst vadose zone behaviour on top of a cave (Bradley et al., 2010; Hartmann et al., 2012b; modified and combined); infiltrating rain water can be stored within the soil (yellow area) for some time, where it is exposed to evapotranspiration. Rainfall exceeding the soil storage capacity enters the epikarst (blue shaded area), which is able to store and redistribute the water laterally from diffuse percolation pathways (small blue arrows) to concentrated percolation pathways (big blue arrows). On its way towards the subsurface, percolating water may enter and leave voids of various volumes, form speleothems, and change its hydrochemical or isotopic composition (see Figs. 3 and 4).

concentration a function of production and loss by upward and downward diffusion (Pumpanen et al., 2003). Gaseous CO_2 is soluble in soil water, forming a weak acid solution that can participate in mineral weathering reactions. Within karst systems, the most common mineral is likely to be carbonate, such as limestone, present from weathering of the underlying bedrock. Due to the high solubility of limestone, soil water is therefore likely to quickly reach carbonate system equilibrium for a given soil air CO_2. The timescale for equilibrium to be reached will vary from minutes to many hours, depending on lithology and rock-water contact time (Svensson and Dreybrodt, 1992). The chemical fate of this soil water depends on its subsequent hydrological evolution. Percolation from the soil reaches the underlying epikarst. This recharge water can be conceptualised as an unknown mixture of two end-members (1) soil water which has equilibrated with soil air CO_2, and (2) rain water that has bypassed the soil store through macropore or other preferential flow mechanisms, with the composition of the rain.

Water percolating into the epikarst will continue to evolve geochemically. The unsaturated zone is a further source of gaseous CO_2, produced by root respiration and microbial activity (Fig. 3). There is increasing evidence that vadose zone CO_2 can be higher than soil CO_2, and infiltration water is therefore likely to continue to gain CO_2 and

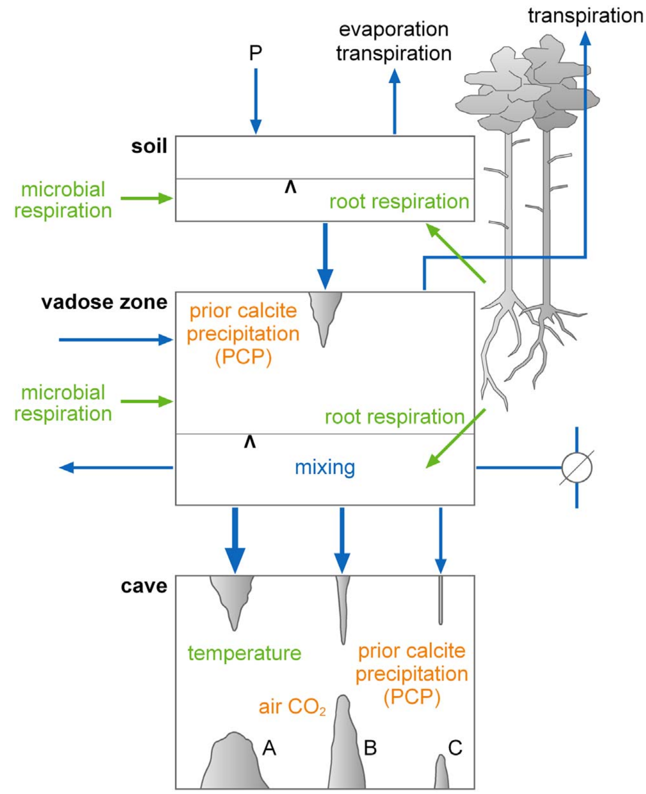

Fig. 3. Processes affecting the growth rate of speleothems. Processes labelled in orange decrease growth rate; processes labelled in green increase growth rate. Speleothem A has the highest discharge (Q) and speleothem C the lowest discharge; ˆ indicates the degree of storage saturation within the soil and the vadose zone.

dissolve more limestone (Baker et al., 2016; Mattey et al., 2016). Within the epikarst and the unsaturated zone, the infiltration water is likely to mix with existing water that is present in dissolutionally enhanced fractures, solution pockets and proto-caves, which can be seen as stores that comprise variable volumes of water and headspace (Benavente et al., 2010). Depending on the shape of the store, the water and air content, and the CO_2 concentrations of both water and air, further carbonate hydrochemical system evolution is likely to occur. Where ventilation occurs, the headspace CO_2 concentration may be less than that of the water, precipitation of calcite will occur (a process widely conceptualised as prior calcite precipitation PCP, Fairchild and Treble, 2009; or prior aragonite precipitation PAP). Depending on the subsequent hydrological and chemical evolution of the water, PCP is likely to decrease drip water calcium concentration and decrease speleothem growth rate. Tree water uptake can cause the concentration, and precipitation, of residual solutes. Through root respiration, trees can add CO_2 to depth in the epikarst and unsaturated zone (Fig. 3), which is mostly the case for shallow caves. Tree mortality can provide another non-linearity in the karst system (Nagra et al., 2016).

Water that finally reaches the cave experiences a highly variable hydrochemical evolution, primarily depending on the drip or seepage water flux into the cave, and the water flow path from the point of ingress into the cave. Speleothem growth rate will primarily depend on the water flux, as well as the relative carbon dioxide saturation of the water compared to the carbon dioxide saturation of the cave atmosphere (Buhmann and Dreybrodt, 1985; Dreybrodt, 1999). The carbonate saturation of the water in the water film on the stalagmite cap will depend on the amount of time the water has already been exposed to the cave atmosphere and undergone PCP in the cave (Fig. 3). One end-member of this process is a complete PCP while the water is still on the stalactite and hence leading to no stalagmite formation. Its other end-member is very rapid water dripping onto the stalagmite every few seconds, in which case a high drip water supersaturation on the stalagmite top is maintained. Most importantly, the formation of speleothem calcite is not an instantaneous process: after the very rapid process of the degassing of CO_2 from the solution, the rest of the reactions take several hundreds of seconds to reach equilibrium (Hansen et al., 2013). The slow process of CO_2 generation from H^+ and HCO_3^-, and diffusion of the CO_2 generated in the thin water film, are both rate-determining (Dreybrodt, 1999). These processes indicate that the half-life of calcite formation τ_p, (i.e. the time taken for half of the possible calcite to form) is of the order of 100s of seconds (Dreybrodt and Scholz, 2011). If drip waters reach the stalagmite surface quicker than τ_p, then it is clear that the reaction will never complete and there will therefore be a disequilibrium reaction. The reasons why this is important are discussed in the following section.

3.3. Transport and fractionation of oxygen isotopes

Other than CO_2 and calcite, whose behaviour is controlled by interactions of rock, water, and vapour, the dynamics of oxygen isotopes are controlled by fractionation during phase transitions and mixing. The volume of soil water may be decreased by evaporation from the soil surface, with the lighter isotopes preferentially evaporated. The extent to which the soil water is evaporatively enriched depends on climatic factors (e.g., temperature, wind speed), as well soil properties, and the time between rainfall events (the latter will dilute any residual enriched soil water). Hence, water reaching the epikarst also has two end-members concerning its isotopic composition: (1) soil water, which has a homogenous isotopic composition that reflects previous rainfall composition and evaporative enrichment, and (2) rain water that has bypassed the soil through macropore or other preferential flow mechanisms with the isotopic of the rain.

Mixing of water in the unsaturated zone storages will change its water isotopic composition in proportion to the volume of stored water and input and output fluxes from the store (Fig. 4). In a simple system, storage and mixing of water can lead to a smoothing of the water isotopic composition. However, the karst is likely to introduce non-linear behaviour, with overflow, flow-switching and nonstationary behaviour all reported, and these can introduce non-linear changes in the water isotope composition (see Subsection 3.1). Additionally, in some circumstances, evaporation of water with associated evaporative fractionation can occur. This is most likely near-surface, where the head-space has a direct connection to the surface.

The amount of water isotope fractionation occurring in the cave is similarly controlled by the amount of time the water is exposed to the cave atmospheric conditions (Fig. 4). For oxygen isotopes, wind speed and relative humidity may be important, as they determine the extent of evaporative fractionation (Deininger et al., 2012; Dreybrodt and Scholz, 2011). The effects of evaporation lead to an increase in abundance of ^{18}O of the H_2O and HCO_3^- in solution. Evaporation is a relatively slow process compared to the time for calcite precipitation (the half-life being τ_p): however, when drip rates are very slow, the isotopic fractionation may be observed in the speleothem calcite (Dreybrodt and Scholz, 2011). More importantly, the composition of the $\delta^{18}O$ within the stalagmite calcite will depend on the extent to which the HCO_3^- in the water film on the stalagmite cap has reached equilibrium, a process that takes several hundred seconds (see Section 3.2). The oxygen isotopes preserved in speleothem calcite are produced by an isotopic fractionation process, as there is a preference for the light ^{16}O in the HCO_3^- to react and convert to $H_2{}^{16}O$, concentrating the amount of the heavy ^{18}O in the remaining HCO_3^- solution. Therefore, the speleothem calcite being produced from an individual drip will become progressively enriched in ^{18}O compared to the solution, and this enrichment only stops when the drip is replaced by the next water drop (Dreybrodt and Scholz, 2011; Fairchild and Baker, 2012; Hendy, 1971; Lachniet, 2009). Speleothem $\delta^{18}O$ is therefore only similar to the initial drip water composition when drip rates are fast (replacing the drop and

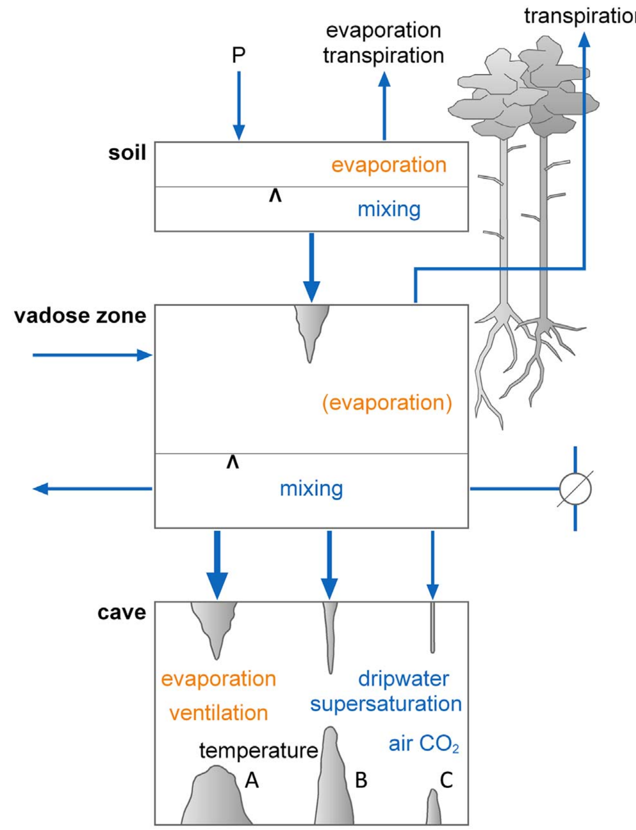

Fig. 4. Processes affecting oxygen isotopic composition of speleothems from atmosphere to cave. Processes labelled in orange enrich the solution with the heavier isotope ^{18}O. Speleothem discharge as in Fig. 4; ^ indicates the degree of storage saturation within the soil and the vadose zone.

associated HCO_3^-), and when HCO_3^- saturation is low (slowing the reaction rate). Finally, there is the case of very slow drip rates, where the water on the stalagmite cap approaches equilibrium, and equilibration of the water in the thin film can occur with the water vapour in the cave atmosphere, leading to the possibility that this signal will also be imprinted in the stalagmite $\delta^{18}O$ (and $\delta^{13}C$).

Most importantly, there is a fractionation of oxygen isotopes that occurs during the reaction processes that form speleothem calcite that is temperature dependent. Often, a gradient of 0.24‰/°C is used, which was derived from laboratory experiments of Craig (1965), O'Neil et al. (1969) and Kim and O'Neil (1997). However, higher gradients have been proposed, both from laboratory experiments which investigated relevant growth rates and pH (Dietzel et al., 2009), and from the natural occurrences of speleothem $\delta^{18}O$ where temperature is known (Coplen, 2007), Tremaine et al. 2011). There is some uncertainty over the relevance of the 0.24‰/°C figure to speleothem formation, and this is a current research focus of the speleothem paleoclimate research community. Applying the appropriate (temperature dependent) fractionation factor is essential if one wishes to convert the $\delta^{18}O$ of speleothem calcite to a water equivalent, and vice-versa, and forms the basis for interpreting paleoclimate from the $\delta^{18}O$ of speleothems (see Subsection 4.3).

3.4. Simulation of vadose zone flow and transport

Up to now, modelling attempts to simulate the karstic unsaturated zone dynamics are mostly related to groundwater recharge estimates. Empirical GIS-based methods are developed at locations where recharge estimates were available. They consider spatial information about geology, soil types, vegetation, mean annual precipitation, land use, infiltration land forms and altitude, and have been used in several

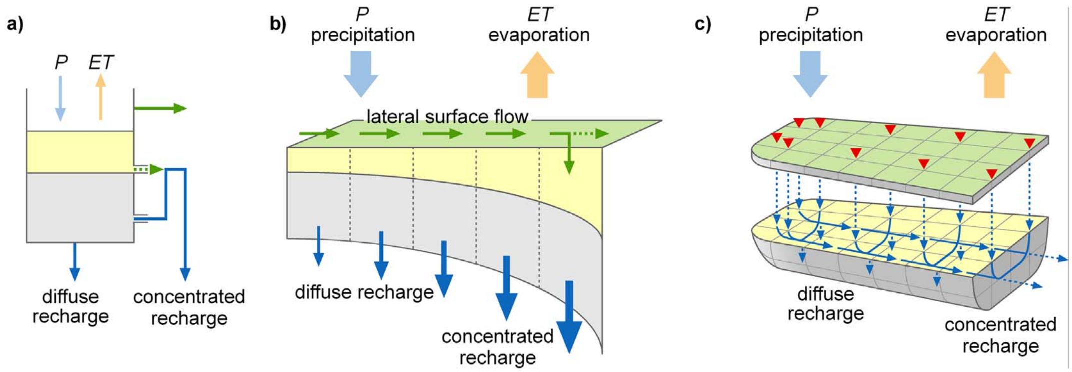

Fig. 5. Different model approaches to simulate the vadose zone: (a) lumped (Tritz et al., 2011; modified), (b) hybrid/semi-distributed (Hartmann et al., 2015; modified), and (c) distributed (Kiraly, 2003, modified).

studies (Allocca et al., 2014; Andreo et al., 2008; Malard et al., 2015b; Radulovic et al., 2012) but their transferability to other regions or towards the future is limited (Hartmann et al., 2014a). Similar to the aquifer scale modelling approaches, karst recharge models can also be classified into lumped and concentrated approaches that consider the unsaturated zone processes by different degrees of complexity.

Lumped recharge estimation approaches (Fig. 5a) were applied in many cases (Fleury et al., 2007; Geyer et al., 2008; Jukic and Denić-Jukić, 2009; Schmidt et al., 2014; Tritz et al., 2011), mostly as a part of aquifer-scale karst simulation of a models at study sites with limited data availability. Other authors relied on physically-based distributed approaches (Fig. 5c, Andreu et al., 2011; Dvory et al., 2016; Hughes et al., 2008; Ireson and Butler, 2011; Kiraly et al., 1995; Martínez-Santos and Andreu, 2010; Perrin et al., 2003) relying on prior knowledge or precise measurements of soil and epikarst hydraulic properties and their spatial variability. Distributed approaches have the advantage of being able to include the spatial variability of recharge while lumped approaches usually provide only one spatially averaged or "effective" time series of recharge and they are not able to include different types of flow paths that were observed at cave drips as mentioned above. Most of these approaches were used to simulate (potential) groundwater recharge, i.e. the water leaving the soil and the epikarst. Only few studies explicitly considered water movement in the vadose zone (Kordilla et al., 2012; Rimmer and Salingar, 2006).

Recently hybrid approaches tried to cope with limited data availability by expressing the spatial variability of soil and epikarst hydraulic properties by (1) representing them by distribution functions (Fig. 5b, Hartmann et al., 2012, 2015) or (2) by taking into account different flow paths and storages within a lumped model (Bradley et al., 2010). These approaches also included solute transport simulations that were based on solute (or isotope) mass balance and the assumption of instantaneous and complete mixing within each of the model reservoirs. Non-conservative behaviour, for instance dissolution of evaporites or decomposition of dissolved organic carbon, were incorporated by conceptual equations (e.g., see Charlier et al., 2012; Hartmann et al., 2014b). Similarly, isotopic fractionation processes have been incorporated in a conceptual way (Baker et al., 2013; Bradley et al., 2010) but it may also be possible considering them by a more physical manner as shown in modelling studies of non-karstic terrains (Schwerdtfeger et al., 2016).

4. Paleoclimate reconstructions by speleothems

4.1. Conceptual model of speleothem growth rate and calcite $\delta^{18}O$ composition

Figs. 3 and 4 show in a simplified manner the processes affecting the two most commonly archived speleothem climate proxies – growth rate and δ^{18}O - by considering three typical speleothem types, defined by their morphology. The morphology is primarily controlled by the water flux, (Q), defined as a drip rate (time between drips) compared to the calcite precipitation rate (τ_{p},; after Buhmann and Dreybrodt, 1985):

A) Speleothem A is a stalagmite boss, formed from a flow regime that has high and constant Q, and therefore just a few seconds between drips (10^0–10^1 s). Water flows down the sides of the stalagmite, forming the low height to width ratio. The stalagmite is paired with a stalactite that has a 'curtain' or 'drapery' shape, with water flowing down the lower surface and precipitating calcite. With respect to the oxygen isotopic composition of the stalagmite, the high Q limits exposure to the cave atmosphere, and therefore fractionations due to evaporation or ventilation is expected to be minimal. The high Q means that the surface water film on the stalagmite cap is replaced with a new drip before much carbonate is precipitated. The water film is therefore maintained at close to its initial HCO_3^- concentration. With respect to stalagmite growth rate, only limited PCP is likely to occur on the stalactite. The fast drip rate means that the water film on the stalagmite is replenished before all the calcite is precipitated, and that this water flows down the sides of the stalagmite, depositing calcite there. The vertical growth rate of this stalagmite will therefore be lower than the maximum possible, and a decrease in Q will increase the vertical growth rate of the stalagmite, as a greater proportion of degassing and calcite precipitation will occur there. Stalagmite growth rate of speleothem A is likely to contain a complex relationship with climate, but δ^{18}O will be most closely related to the δ^{18}O of the drip water.
B) Speleothem B is a columnar stalagmite, with water dripping onto it along the outside of an icicle shaped stalactite. For this pair of speleothems, Q is lower, with the time between drips (10^2 s) similar to that of τ_p. With respect to the stalagmite oxygen isotopic composition, the low Q means that the oxygen atoms in the water molecules can potentially undergo fractionation in the cave for a longer time than in the case of stalagmite A. If the time between drips is similar to τ_p, kinetic fractionation during speleothem calcite precipitation will be maximum. Depending on the cave air relative humidity and wind speed, fractionation may occur due to evaporation, in which case the effect would be considered greater for stalagmite B compared to speleothem A. With respect to growth rate, PCP occurring as the water flows over the stalactite surface will be more significant than at speleothem A, as Q is lower and there is more time for degassing on the stalagmite and calcite precipitation to occur. In contrast, the vertical stalagmite growth rate will be relatively optimal with respect to Q, with no flow down the sides of the speleothem. With the water drop being replaced at a time similar to that τ_p, both drip water carbonate saturation and Q maybe be the limiting factor on growth rate. Similar to speleothem A, stalagmite growth rate is likely to be a complex relationship with climate.
C) Speleothem C is a pair of speleothems where water drips from

within a 'soda straw' stalactite onto a 'candlestick' stalagmite. For this speleothem pair, the time between drip is slower than τ_p, and water supply Q becomes a significant control on the stalagmite hydrochemistry. With respect to oxygen isotopes, the drip water is sourced from within the soda-straw stalactite, so it is protected from both degassing and within-cave evaporation processes until it is exposed to the cave air on the stalactite tip. Here, and on the stalagmite cap, in caves with low relative humidity or high ventilation, the water may undergo fractionation due to the long exposure time to the cave atmosphere. In extreme cases, evaporation may lead to a higher relative abundance of oxygen atoms in the water molecule compared to the abundance of oxygen in carbonate. Complete reaction of HCO_3^- to form speleothem $CaCO_3$ will occur before the next drip occurs, and therefore the maximum amount of possible oxygen isotope fractionation will have occurred, with the extent of fractionation controlled by the initial HCO_3^- concentration of the water on the stalagmite cap. For very slow drips (> 10 min), equilibration of the water film with the cave atmosphere is possible (Dreybrodt and Scholz, 2011). With respect to growth rate, the low Q allows for calcite precipitation (PCP) at the tip of the stalactite forming a 'soda straw' type stalactite. Stalagmite vertical growth rate is now limited by water supply, in contrast to stalagmites A and B, as drip rate is slower than the time taken to precipitate calcium carbonate. However, the $\delta^{18}O$ record in the stalagmite is likely to be one controlled by fractionation processes.

Considering the hydrological controls on speleothem paleoclimate proxies, and the best speleothems to use for climate reconstruction from oxygen isotopes or growth rate, then we can conclude the following: Stalagmite growth rates may be sensitive to drip rate, and therefore rainfall recharge, only for samples which are hydrologically controlled by water limitation (low Q, when the time between drips is great than τ_p). For faster drip rates, stalagmite growth rate is likely to be a complex proxy of cave drip water supersaturation, drip rate and cave air carbon dioxide (Buhmann and Dreybrodt, 1985; Dreybrodt, 1999), and more likely to be climatically ambiguous. And even for water-limited samples, the drip rate control on speleothem growth rate can still be overwritten by variability in cave drip water supersaturation or cave air carbon dioxide. Speleothem $\delta^{18}O$ is likely to be effected by in-cave fractionation processes for all stalagmite types. So, if this effect is desired to be avoided, the sample choice is highly predicated by the choice of the optimal cave climate - ~100% relative humidity and 0 m/s ventilation to avoid evaporation (Hendy, 1971). Alternatively, the extent of disequilibria can be quantified and related to one or more processes, and permit the degree of disequilibrium to be interpreted as a climate proxy. Fast dripping and slowly degassing samples will be least affected by kinetic fractionation (Hendy, 1971). In these samples, $\delta^{18}O$ should be dominated by climatic processes (water $\delta^{18}O$ composition and the temperature-dependent fractionation during the formation of calcite), moderated by karst hydrology.

4.2. Quantifying sources of uncertainty in speleothem $\delta^{18}O$

As speleothem $\delta^{18}O$ is the most widely measured geochemical proxy, here we investigate the sources of uncertainty in climate, vadose zone hydrology and in-cave processes, and quantify their effect on speleothem $\delta^{18}O$. We have provided a theoretical outline of how speleothem $\delta^{18}O$ might be deposited close to equilibrium with respect to their drip waters for speleothems that are fast dripping, slow degassing, in caves with ~100% relative humidity and no air flow (Section 4.1). Models have been developed that quantify the extent to which the speleothem $\delta^{18}O$ might be affected by these disequilibrium and fractionation processes (Deininger et al., 2012; Dreybrodt and Scholz, 2011). These show that speleothems deposited out of equilibrium with respect to $\delta^{18}O$ will have a 'worst case' composition that differs by up to ~3‰ from the drip water value (Deininger et al., 2012). These models compare favourably with empirical evidence of $\delta^{18}O$ isotopic disequilibria in speleothems from laboratory analogue experiments (Day and Henderson, 2011; Fantidis and Ehhalt, 1970) and comparison of modern day drip waters and 'farmed' calcite grown on glass plates (Mickler et al., 2004, 2006).

Assuming speleothem samples are chosen that are likely to be deposited close to isotopic equilibrium with the drip waters, the question then arises: what are the ideal regions and time periods where drip water $\delta^{18}O$ represents a useful climate or environmental parameter such as temperature, rainfall, climatic seasonality, evaporation or groundwater recharge? Modern cave monitoring studies provide a useful insight into regional patterns in drip water $\delta^{18}O$. For example, these can be used to examine the extent to which drip water $\delta^{18}O$ reflects the weighted mean of precipitation $\delta^{18}O$. Monitoring studies in cool and temperate climates demonstrate that $\delta^{18}O$ values of drip water and $\delta^{18}O$ values of precipitation are in reasonable agreement: e.g., < 0.3‰ difference in Scotland (Fuller et al., 2008) and France (Genty et al., 2014). In contrast, in water-limited environments, $\delta^{18}O$ drip water has been reported to be variable between drips within a cave site (e.g., ~2‰ between mean drip water compositions in an Australian cave, Cuthbert et al., 2014), and both substantially lighter (up to −1‰, Texas, Pape et al., 2010) or heavier (up to +2.8‰, Cuthbert et al., 2014) than the weighted mean of precipitation. In water limited environments, variability in $\delta^{18}O$ is increased due to (1) the greater potential for evaporative fractionation of water isotopes in the soil and shallow karst; (2) relatively empty karst fractures and stores, with less mixing of waters between events, increasing hydrological variability and non-linearity; and (3) recharge more likely from long-lasting or intense rainfall events that exceed the soil moisture capacity, biasing recharge water to more negative $\delta^{18}O$ values.

Modern cave monitoring studies therefore suggest that drip water $\delta^{18}O$ is most likely to be less variable between drips, and more likely to directly record the weighted mean $\delta^{18}O$ of precipitation, in regions with water excess (P > E) for most or all of the months of the year, which limits the opportunity for evaporative fractionation and permits mixing of recharge water in karst stores and fractures. However, the $\delta^{18}O$ of precipitation is not necessarily a sensitive indicator of climate or recharge processes in all regions of the world. Local precipitation $\delta^{18}O$ sampling campaigns, the Global Network of Isotopes in Precipitation (GNIP) database, and isotope-enabled climate model output can all be used to identify suitable regions where $\delta^{18}O$ of precipitation is climatically sensitive (e.g., Bowen, 2008). For example, eastern Europe is at the limit of the influence of westerly airstream associated with the North Atlantic Oscillation (NAO), and in Germany winter $\delta^{18}O$ is +1.8‰ higher in positive NAO years (Baldini et al., 2008). In SE Asia, Moerman et al. (2013) showed that inter-annual variations in $\delta^{18}O$ of precipitation in Mulu are 6–8‰, explaining 70% of the variance in $\delta^{18}O$ and correlating with both local precipitation amount and ENSO.

In this section, we have used empirical evidence to quantify the sources of and extent of uncertainty in $\delta^{18}O$. Some regions are 'hotspots' - where precipitation $\delta^{18}O$ is variable and sensitive to a climate parameter, and such regions are more likely to provide speleothem $\delta^{18}O$ where the climate 'signal' dominates over any hydrological or geochemical 'noise'. For example, there is widespread success in speleothem $\delta^{18}O$ recording global climate change at the timescale of glacial-interglacial cycles from the monsoonal and low-latitude climate zones (Carolin et al., 2016; Cheng et al., 2016; Wang et al., 2017; Wang, 2001), with a typical change in speleothem $\delta^{18}O$ of 2–4‰ from glacial to interglacial state and ~1–2‰ between interstadials. This demonstrates the dominance of climate forcing on speleothem $\delta^{18}O$ over this timescale and amplitude of global change, rather than hydrological and geochemical uncertainties that originate from the karst system (Fairchild et al., 2006). Changes in precipitation amount, moisture source region and temperature all combine to generate a reproducible speleothem $\delta^{18}O$ signal that dominates other sources of uncertainty. In contrast, speleothem $\delta^{18}O$ for the same time period from mid-latitude sites have a $\delta^{18}O$ signal that can be more ambiguous (e.g., < 2‰ from glacial to interglacial state (Hellstrom et al., 1998; Williams et al., 2015) and < 1‰ difference between interstadial events; Genty et al., 2003). In these regions, oxygen isotopic response to changes in moisture source region or precipitation amount may be comparative small, or act in the opposite direction temperature effects.

Over periods of relatively stable climate, and smaller changes in climate forcing, interpreting individual speleothem $\delta^{18}O$ becomes harder, as the climate 'signal' becomes harder to distinguish from any hydrological or geochemical 'noise'. This 'noise' is greatest in water-limited environments, where drip water $\delta^{18}O$ can be both offset from the weighted mean of precipitation (< 3‰), and show high variability between drips (< 2‰). In addition, in any region it is possible for some speleothems to form with a significant extent of non-equilibrium isotope deposition, with $\delta^{18}O$ offset by < 3‰. For example, a comparison of European Holocene speleothem $\delta^{18}O$ successfully demonstrated a ~0.13‰ / per degree longitude trend in speleothem $\delta^{18}O$: however, mean speleothem $\delta^{18}O$ is offset by 1‰ due to non-equilibrium deposition (McDermott et al., 2011). For shorter time periods, such as the climate of the last two thousand years, the challenge in quantifying the relationship between speleothem $\delta^{18}O$ and climate and hydrology is significant. We propose that a vadose zone modelling approach helps understand and constrain the processes determining both mean values and uncertainties in speleothem $\delta^{18}O$ over all time periods.

4.3. General principles for paleoclimate reconstructions by speleothems and research gaps

We have previously demonstrated the heterogeneity of karst hydrology (Sections 2 and 3), which leads to a diverse range in drip water discharge responses (Q) to a specific climate input. In addition, we have shown that the two most widely used climate proxies used in cave speleothems, oxygen isotopes and growth rate, have the best hydroclimate sensitivity in completely contrasting speleothem types (Subsection 4.1). Speleothem $\delta^{18}O$ is most similar to drip water $\delta^{18}O$ in samples with high drip discharge Q (the time between drips is less than τ_p), and growth rate has the simplest relationship with water availability with low Q (the time between drips is greater than τ_p). However, the past drip discharge in a fossil speleothem sample is almost impossible to determine. Empirical evidence (Baker and Smart, 1995) suggests a drip rate of $1 \times 10^{-5}\,L\,s^{-1}$ (~1 drip every 15 s) or faster will lead to HCO_3^- saturated water flowing down the sides of a speleothem and forming speleothems with increasing width to height ratio, or flowstones. For columnar or candlestick-shaped stalagmites, which are no longer forming, determining the past drip rate is impossible, and we are reliant on the proxy evidence (e.g., $\delta^{18}O$, growth rate) to derive a record of past recharge.

In order to obtain a record of past hydroclimate from speleothems, four research tasks should be undertaken. Firstly, the monitoring of the modern-day cave hydroclimate and modern calcite deposits helps understand modern-day karst hydrology, drip water geochemistry and cave climatology. As well as providing a modern-day understanding of the karst system, this data can be used as input variables into a forward model (or proxy system model, Evans et al., 2013). Secondly, the forward model can be used to provide a process-based understanding of speleothem paleoclimate proxies. These permit the quantification of the uncertainties associated with the processes affecting the geochemical proxy, and they can be used to produce modelled proxy time series (or pseudoproxies), which can be compared to actual speleothem data. Thirdly, where possible, speleothem proxy data for the period of instrumental data can be correlated to measured climate variables. This provides empirical evidence for climate relationships that were identified by the forward model and, in some instances, provides a quantification of a climate record. Finally, the replication of stalagmites helps determine when climate variables are dominant over other non-climate processes e.g., non-linear hydrology or kinetic fractionation.

Within these general principles, a few recent advances and research gaps can be highlighted.

1) Modern monitoring of cave processes has been advanced by the availability of automated loggers for crucial parameters such as drip rate (Collister and Mattey, 2008; Luetscher and Ziegler, 2012), cave air CO_2 concentration, and temperature. The measurement of drip water geochemistry will remain a specific challenge, as waters need to be collected and preserved as rapidly as possible to preserve the original water chemistry. In remote sites or caves that are difficult to access, obtaining comprehensive hydrochemical datasets is unlikely.
2) In general, cave climate is poorly monitored in comparison to drip water hydrology and chemistry. This is important given the potential effect of air currents and relative humidity on the extent of disequilibrium oxygen isotope fractionation, and the temperature dependence of oxygen isotope incorporation into calcite. Automated probes that can measure relative humidity in high humidity environments and wind speed at low wind speeds are available, but rarely used. Challenges include the accurate and precise measurement of very low wind velocities, and the accurate measurement of relative humidity in high humidity cave environments. The latter requires expensive and energy-demanding relative humidity probes that contain heaters to remove condensation. Cave air and drip water temperature are also rarely measured, with the assumption made that cave air and water temperature is the same as the external surface air temperature. Numerous studies have now shown this assumption to not be true (Domínguez-Villar et al., 2013, 2015; Rau et al., 2015).
3) Modern monitoring of surface hydroclimate is also highly informative. Most poorly constrained is tree water use (transpiration) as part of the water balance Measurement of tree water use is a challenge, as is the case for groundwater research in general. The estimation of evapotranspiration typically relies on regional hydrological products. Soil water content is similarly important to understand recharge thresholds, yet soil moisture probes are rarely installed in monitoring programs. An ideal surface monitoring program would include a weather station equipped to measure all standard meteorological parameters, as well as actual evaporation, soil moisture and soil carbon dioxide, and monitoring of tree water use using sap flow meters.
4) Modern monitoring of rainfall $\delta^{18}O$ is essential to understand speleothem $\delta^{18}O$, and in many cases the challenge is how to collect rainfall samples close to a remote cave site. Monthly integrated samples using IAEA standard protocols allow comparison with the IAEA rainfall isotope monitoring network, which is very sparsely distributed globally. This sampling is relatively easy to maintain at sites that can be visited monthly, however monthly integrated samples are highly unlikely to reflect the actual $\delta^{18}O$ of rainfall that generates recharge. Daily samples, grab samples and within-event samples are all valuable in this regard, many of which can be successfully collected and stored by community-based volunteers as well as trained scientists. An increasing number of satellite-based isotope products are becoming available: these will be valuable but also require local benchmarking of their integrated signal of water in the atmosphere by observed isotope data.
5) There is a need for water isotope time-series from isotope enabled global climate models that have been run over past climate periods. Although model output from multiple models does exist for some time periods (e.g., mid Holocene, Last Glacial Maximum) arising from research community programs such as the Paleoclimate Model Inter-comparison Projects such as PMIP2 (Braconnot et al., 2007), longer time-series would be valuable. The limitations to such data becoming available include the incorporation of water isotopes into climate model code, and the computational time cost it takes to run long climate simulations. With increasing refinement of land-surface models, useful products could include multi-millennial soil and

groundwater $\delta^{18}O$ as well as precipitation $\delta^{18}O$ data that can be used to constrain forward models.

6) Replication is a crucial aspect of science, and replication of speleothem records is important given the possibility of non-linear responses in karst hydrology to climate, and its effect on speleothem proxies. The particular challenge here is the time and analytical cost of replication of speleothem proxy records, especially if sampled at high temporal resolution. This suggests a community focus on a limited number of climatically sensitive karst sites would be productive.
7) Forward modelling of speleothem paleoclimate proxies is a relative new approach, and one that has been developed independently in numerous paleoclimate research communities over a similar time period. For a review of forward models, in the wider community also known as proxy system models (PSMs), we refer to Evans et al. (2013), with implementation of a forward modelling package for multiple proxy archives (PRYSM) found in Dee et al. (2015). The development of karst hydrological models, and their use as forward models or PSMs is covered in detail in Section 5.

5. Karst hydrology models and paleoclimate reconstruction

Although the processes determining the $\delta^{18}O$ composition of speleothem drip waters, the associated speleothems and their growth rate are increasingly well understood, sources of uncertainty remain (Subsection 4.3). Many of them are related to the hydrology and related transport processes within the vadose zone (see Sections 2 and 3). For this reason simple karst hydrological models have been introduced to reduce the uncertainty (Baker et al., 2013; Bradley et al., 2010). Based on the principles provided in the previous chapters, we propose further improvements in the representation of hydrological and hydrochemical processes in these models. We see notable potential in the following topics:

5.1. Modelling of evapotranspiration processes

5.1.1. Evaporation in the soil and epikarst

Evaporation in the soil and epikarst is a significant process, affecting oxygen isotopic composition, and the water balance. Drip waters which are isotopically heavier than the weighted mean of precipitation have been observed in arid and temperate semi-arid regions (Ayalon et al., 1998; Cuthbert et al., 2014), as well as in a transect of sites across China (Duan et al., 2016). Changes in stalagmite $\delta^{18}O$ in the mid-Holocene in Midwest USA have been interpreted as changes in the amount of evaporative enrichment occurring as vegetation changed from prairie to forest (Denniston et al., 1999); evaporative enrichment of drip waters due to loss of canopy covers and shading have also been observed after wildfire (Nagra et al., 2016). The effect of evaporative fractionation of karstic vadose zone waters depends on the intensity of evaporation; the amount and frequency of diffuse recharge (which may mix with any residual, isotopically enriched soil or epikarst water and transport it to a speleothem forming drip), and the karst architecture (e.g., depth of stored water and its connectivity to the surface). With respect to existing karst hydrological models, existing models parameterise the soil and epikarst evaporative fractionation as a poorly constrained rate term (Bradley et al., 2010; Cuthbert et al., 2014). This is due to the paucity of empirical data that is available to constrain the rate of evaporative fractionation in both soil water and epikarst water. For example, soil water samples can be difficult to obtain where there are thin karst soils, and epikarst water sampling requires specialist shallow piezometer installations.

5.1.2. The role of tree water use

The depths below surface that trees access groundwater, and how that varies with species and with climate and other environmental factors is a subject of considerable uncertainty. Direct hydrological observations of tree water-use on speleothem-forming drip waters are rare (Coleborn et al., 2016, being an exception), despite the fact that tree roots are often observed penetrating into and past caves at depths of up to at least 10 m below the surface, with root respiration providing a source of carbon dioxide for carbonate dissolution. Within karst hydrological models, tree water use is rarely considered. To our knowledge, Sarrazin et al. (2016) were the first to include a vegetation parameterisation in a karst recharge model. Such inclusion of transpirative water loss by trees and vegetation within karst hydrological models would permit the modelling of changes in water balance over time due to forest growth or land cover changes, and the effect on water balance and drip water calcite supersaturation.

5.1.3. Within-cave oxygen isotope fractionation

At the time of writing, two approaches to modelling in-cave oxygen isotope fractionation processes were available. Theoretical approaches presented by Dreybrodt (2008) and Scholz et al. (2009) are partially reconciled in Dreybrodt and Scholz (2011), resulting in a MATLAB model of in-cave oxygen isotope fractionation (iSOLUTION) presented in Deininger et al. (2012). A critical limitation of all the models is the lack of field validation of the model structure and their parameterization. To date, no cave site has been monitored for all the processes that can determine the extent of oxygen isotope fractionation (wind speed, relative humidity, drip rate, drip water supersaturation, cave air carbon dioxide). This is a critical area for future research, and recent technological advances in cave monitoring now make the automated, in-situ monitoring of these parameters possible (see Section 5.3).

5.2. Selection of adequate model structures

5.2.1. Simulation of individual cave drip dynamics

Considering the previous subsection, it is becoming obvious that there is still a high potential to reduce uncertainty due to vadose zone evapotranspiration issues by process-based modelling. However, integrating these processes in a complex vadose zone hydrology and transport model will require detailed knowledge about the parameters that control vadose zone hydrology and transport.

Simple single-reservoir models of drip water oxygen isotope composition (e.g., Baker and Bradley, 2010; Truebe et al., 2010; Dee et al., 2015) were not able to fit sites where there is observed non-linearity in karst drip water hydrology but they were computationally efficient and there were few model parameters to be identified. In contrast, more detailed lumped parameter models such as that of Bradley et al. (2010) are more process-based and more adequate for a complex karstic vadose zone. However, in terms of parameter identification they may be considered to be over-complex since observations to facilitate model parameter identification are limited. Karst hydrological models are usually evaluated by their performance, the identifiability of their model parameters and the realism of their simulations (Hartmann et al., 2013b). But such thorough evaluation is only possible if long time series of drip observations in a high resolution (days or less) are available. If data availability is reduced to the temporal resolution of speleothem analyses (seasonal, years or even larger) such evaluation is hardly feasible.

Based on a 8-year data set of drip rate and a hydrochemistry monitoring programme, Treble et al. (2015) demonstrated that changes in model configuration over time are needed to fit observed drip water hydrology and oxygen isotope data at a cave site in Western Australia. The data that was collected in this intense monitoring programme allowed applying a more complex and more realistic model and its evaluation. If used for paleoclimate reconstruction at the same site, the model's skill in reducing uncertainty due to hydrological processes would be enhanced compared to sites where no intense monitoring programme was used to understand and parametrize vadose zone processes. By developing and applying as semi-distributed karst vadose zone model simultaneously to a set of drip rate observations, Hartmann et al. (2012) could adequately reproduce short-term and seasonal drip rate and solute transport at a karst cave in northern Israel. Applying an elaborated parameter estimation procedure, they could identify the information provided by the different observations and quantify the remaining uncertainty.

We believe that at the scale of an individual cave, such approaches that monitor the present hydrological behaviour provide great potential to quantify the impact of vadose zone hydrology on paleoclimate reconstruction. Detailed monitoring programs of drip rates and their hydrochemical composition (including $\delta^{18}O$) will facilitate the development of site-specific process-based model structures that can be later applied for paleoclimate reconstruction during time periods much earlier than the monitoring period.

5.2.2. Simulation of drip dynamics at ungauged caves

In many cases, $\delta^{18}O$ records of speleothem composition are available without a detailed monitoring of drip rates and their hydrochemical composition. Hence, the choice of the "most adequate" model structure and a proper choice of model parameters is not directly possible. In such cases, modelling and parameter estimation has to rely on a-priori understanding of vadose zone hydrology and its incorporation in a vadose zone model (Section 3). Such an approach was pursued by Bradley et al. (2010), who used a lumped model that expresses the typical flow paths and storages of the karst system on top of the cave with an a-priori parameterization. Recent $\delta^{18}O$ records of the speleothems allowed evaluating the adequacy of the model.

Another possibility to include the hydrology of the vadose zone is the use of recharge simulations derived from large-scale hydrological models (e.g., Hempel et al., 2013), which are usually parametrized using a-priori information. However, operating on a coarse global grid (often 0.25–0.5 decimal degree), these models often fail to reproduce the spatiotemporal variability of karst recharge processes (Hartmann et al., 2017). Karst-specific large-scale recharge simulation approaches (e.g., Hartmann et al., 2015; Rahman and Rosolem, 2017) may provide more realistic simulations but have to be tested for their performance to reproduce drip rates and drip hydrochemical compositions before they can be applied to ungauged caves.

5.2.3. Simulation of cave drip dynamics as part of entire karst systems

Catchment scale karst models (Subsections 2.2 and 2.3) can take advantage of a wider range of information as they are able to include observations of groundwater levels (Doummar et al., 2012; Reimann et al., 2014), spring discharge (Fleury et al., 2009; Rimmer and Salingar, 2006), water quality data (Charlier et al., 2012; Hartmann et al., 2013a), or artificial tracer information (Oehlmann et al., 2014). Many of these models include subroutines to estimate groundwater recharge and they would therefore provide another opportunity to obtain hydrological simulations for speleothem analysis. However, field studies showed that these average or "effective" groundwater recharge rates and dynamics may not correspond directly to the observed drip rates in a cave (Lange et al., 2010; Mahmud et al., 2016). More detailed field campaigns and modelling studies show that the behaviour of several cave drips within a cave can only be modelled if a range of possible flow paths with varying soil and epikarst thicknesses as well as varying hydraulic conductivities are considered (e.g., Arbel et al., 2010).

Consequently, there is a need to better understand the relation between the drip dynamics within a cave and the behaviour of the entire karst system that surrounds the cave. We believe that using study sites with nested information, i.e. soil moisture observations, cave drip observations, and groundwater level observations at the plot scale, and spring discharge observations that express the dynamics of the entire karst system is a promising direction to understand the relation between cave and karst system hydrology. Model structures that are available to reproduce this relation could be parameterized with karst spring discharge observations and could be applied to cave drips within the karst system. If applied for paleo-reconstruction the integrated relationship between cave drip dynamics and karst system dynamics of these models could also be used to provide estimates of paleohydrology at the scale of the entire karst system.

5.2.4. Non-stationarities of karst system properties

One of the greatest challenges in using karst models for speleothem reconstruction is the non-stationarity of karst system properties. Because caves, as well as their inflow pathways evolve by carbonate rock dissolution (Subsection 2.1), their hydraulic properties can be expected to change over time. Hence, model structures and their parameters have to be assumed non-stationary, too, in order to provide reliable simulations of the vadose zone hydrodynamics for time periods that lie far in the past. One way to cope with this is focussing only on time periods not too far in the past, for which karstification processes can be neglected (a few thousand years). If time periods beyond are to be considered, karst evolution approaches have to be applied. Preceding studies (Bloomfield et al., 2005; Hubinger and Birk, 2011; Perne et al., 2014) showed that, given information about initial porosity, fractures, and age of the karst system, the evolution of structural karst system properties can be assessed by physical and chemical laws. Such approaches could provide a conceptual basis to assess the change of hydraulic parameters that control drip dynamics (Kaufmann, 2003) but they have to be simplified in order to be incorporated in a hydrological modelling approach (e.g., Malard et al., 2015a).

5.3. Linking hydrology and speleothem composition for advanced paleoclimate reconstruction

Despite the complexity of the speleothem $\delta^{18}O$ record, this paleoclimate proxy has been used to reconstruct past water availability. For example, despite speleothem calcite having values 0.3 to 1.2‰ higher than expected for the modern-day temperature and rainfall $\delta^{18}O$, Lachniet et al. (2004) were able to reconstruct 1500 years of tropical rainfall history at the Isthmus of Panama in terms of relatively 'wet' and 'dry' periods. For this study, there was no speleothem deposition over the period of instrumental data, and calibration against modern climate data was not possible. A hydrological model would therefore help constrain the uncertainties associated with speleothem $\delta^{18}O$ for this sample: the timing of rainfall recharge, regional variations in rainfall $\delta^{18}O$ and non-equilibrium deposition (Lachniet et al., 2004). For a similar historic time period, Medina-Elizalde et al. (2010) investigated the $\delta^{18}O$ record from a speleothem from the Yucatán, Mexico. For this sample, instrumental calibration of speleothem $\delta^{18}O$ and modern rainfall was possible, and this permitted the authors to identify drought periods over the last 1500 years, and relate these to the decline of the Maya civilisation. A comparison of the speleothem $\delta^{18}O$ record with regional lake sediment $\delta^{18}O$ records, combined with a hydrological and $\delta^{18}O$ mass balance model, allowed Medina-Elizalde and Rohling (2012) to show that the collapse of the classic Maya civilisation was probably due to a modest reduction of summer precipitation. Finally, Jex et al. (2011) were able to reconstruct autumn-winter precipitation in North-East Turkey from speleothem $\delta^{18}O$ for the last 500 years. By applying an empirical calibration against modern rainfall data, speleothem $\delta^{18}O$ was shown to relate to the North Sea Caspian climate pattern. Subsequent use of a hydrological model (Jex et al., 2013) permitted the investigation of potential non-stationarity in this climatic relationship, and the effects of routing of water through the karst aquifer.

In these studies, the researchers were able to establish a relationship between rainfall amount and $\delta^{18}O$ composition of the rain with different degrees of quantification and uncertainty. The assumption that vadose zone hydrology (Section 3) did not affect the isotopic signal of the percolating waters could be tested through the use of hydrological modelling (e.g., Jex et al., 2013). To elaborate the importance of developing karst hydrological models for speleothem proxy climate records, we take the example of $\delta^{18}O$ and growth rate records from

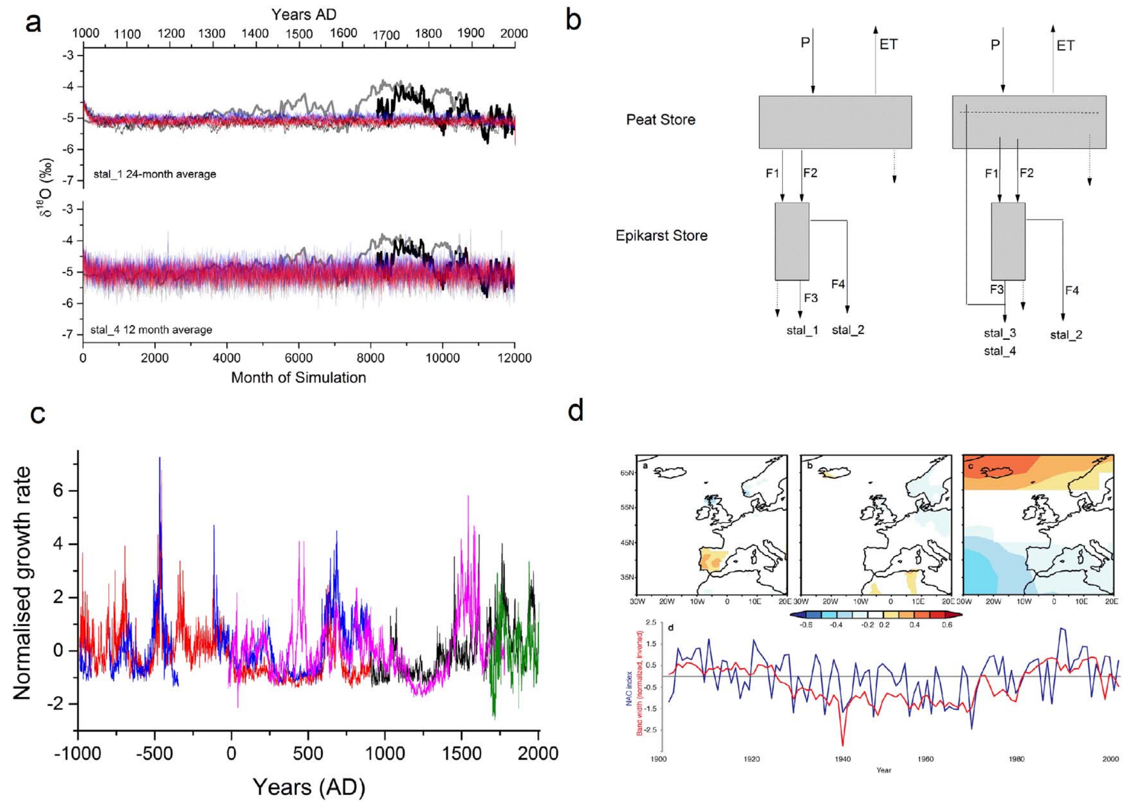

Fig. 6. (a) $\delta^{18}O$ from two stalagmites (Baker et al., 2012), shown in thick grey and black lines, compared to pseudoproxy stalagmite series generated by the two hydrological model structures shown in (b). Different colour pseudoproxy series (thin red, blue and black lines) represent different climate input data. Pseudoproxy outputs are shown for stal_1 (no preferential flow model) and stal_4 (preferential flow model). The different smoothing used reflects the sampling resolution of speleothem calcite. (b) Two alternative karst hydrological model structures for this site. P and ET are total monthly precipitation and evapotranspiration respectively, and F1 to F4 are hypothetical fluxes between the two karst stores Note the large peat water store compared to the epikarst store, reflecting the known modern-day hydrology. (c) Five stalagmite growth-rate series. From the same cave, each colour represents a different stalagmite. Black and green stalagmites are the same as shown in (a). The growth rate data is normalised as individual samples have different mean growth rates, due to local variations in drip water HCO_3^-. (d) Correlation of speleothem growth rate over the instrumental period with climate data. Growth rate data is normalised and labelled band width. Insets show the spatial correlation with (a) winter rainfall (b) temperature (c) sea level pressure and (d) the winter North Atlantic Oscillation (Baker et al., 2015).

stalagmites at one cave in NW Scotland.

At this site, oxygen isotopic composition of two annually laminated stalagmites that have deposited within the last 1000 years have been compared to modern cave monitoring data (Fuller et al., 2008), instrumental climate data including GCM rainfall (Baker et al., 2011), and a forward model of stalagmite $\delta^{18}O$ (Baker et al., 2012). The stalagmite and forward model $\delta^{18}O$ time series are presented in Fig. 6a, and the hydrological structure of the model in Fig. 6b. Drip water $\delta^{18}O$ was identical to the weighted mean of precipitation (Fuller et al., 2008), suggesting that the speleothems in the cave could preserve a record of precipitation $\delta^{18}O$ and climate. However, monitoring demonstrated that drip rates had a very low Q (several minutes or lower between drips), much longer than τ_p, and that the stalagmites are not ideal for $\delta^{18}O$ paleoclimate reconstructions as they are likely to have undergone kinetic fractionation. However, this is mitigated somewhat by the drip waters having low HCO_3^-.

Comparison of the stalagmite $\delta^{18}O$ over the instrumental period demonstrated no correlations between stalagmite $\delta^{18}O$ and any climate fields (Baker et al., 2011). A forward model was developed which represented the karst hydrology of the site, which comprised a large peat water store and a shallow, fractured marble aquifer, with no primary porosity (Fig. 6b). Two alternative model structures were used, permitting the modelling of preferential water flow through the peat. Observational evidence of peat cracks suggested that the direct routing of surface precipitation to the limestone is possible. The model validation had to permit annual water fluxes from the peat store, necessary for the formation of annual laminae from fluorescent organic matter, and the continuous drip water supply for hundreds of years. Over the last millennium, application of both forward models (Fig. 6a) identified the period of 1620 to 1880 CE where the stalagmite $\delta^{18}O$ fell outside the range modelled using modern climate variability and all plausible karst hydrology scenarios (Baker et al., 2012). Whether this is a climate or fractionation signal, e.g., equilibration with cave air, could not be elucidated. Further replication of stalagmite $\delta^{18}O$ records from the cave is necessary to resolve this question.

In contrast, a replicated stalagmite annual growth rate record from Uamh an Tartair, Scotland, comprises five annually laminated stalagmites which have grown over various periods over the last 3000 years (Fig. 6c). This includes the two samples with $\delta^{18}O$ records. These were actively growing over the instrumental period, permitting the calibration of the growth rate record against instrumental climate data (Fig. 6d). This calibration demonstrated a relationship between growth rate, mean annual precipitation, and the winter North Atlantic Oscillation, with slower speleothem growth in wetter conditions and a positive NAO mode (Baker et al., 2015; Trouet et al., 2009). The inverse relationship between growth rate and surface P could be considered counter-intuitive, as it could be expected that wetter conditions would increase the supply of HCO_3^- saturated drip waters, and therefore increase growth rate. Ten years of cave monitoring data demonstrates the stalagmite forming drips have very low drip rates (Fuller et al., 2008), and therefore water supply should be a limiting factor for speleothem growth. However, drip water calcium concentrations were also variable over time (in the range 30–80 mg/L). Cave drip water monitoring in 1997, after six years of persistently positive winter NAO, showed that drip water calcium concentrations in the cave were between 30 and 40 mg/L, barely above saturation, whereas monitoring in 2004, after several years of variable NAO, yielded concentrations in the range 45–60 mg/L for the same drip waters. Drip water HCO_3^- saturation therefore varies significantly over time at this site. We hypothesise the water balance control on soil and vadose zone CO_2 production, where wetter conditions lead to saturated peat and low CO_2 production, and controlling drip water HCO_3^-, is the dominant control on growth rate. Development of a growth rate forward model (Fig. 4) would be helpful in this regard, which should use the same hydrological structure as that for speleothem $\delta^{18}O$ at the site (Fig. 6b). This is the focus of current research.

6. Conclusions

This review paper established the link between paleoclimate reconstruction and karst vadose zone hydrology. It points out that karst hydrological processes within the vadose zone can result in increased uncertainty when speleothems are used for paleoclimate reconstruction. Modelling approaches from karst hydrogeology can be incorporated in paleoclimate reconstruction to reduce uncertainty but, in order to be successful, these models have to comprise the dominant processes controlling the flow and transport dynamics in the vadose zone of the investigated cave. Up to now, many processes that were identified by experimental research have not been incorporated into vadose zone karst models because of problems of model parameter identifiability.

Hence, our recommendations for future research are to gain a better understanding of processes that control vadose zone flow and transport processes and to develop approaches to incorporate these processes into simulation models including methods to identify realistic model parameters. Finally, the most exciting direction that we see is the challenge of reconstructing paleohydrology, which we believe may be achieved using karst hydrology and transport models combined with speleothem composition and speleothem growth analysis. If we can use investigations of paleo-signals from caves to draw inferences about the dynamics of entire aquifers or catchments, we may be able to understand how hydrological systems behaved, as a result of all possible feedback mechanisms, in the past when other climatic periods prevailed. It would therefore contribute to our general understanding of how possible future climatic changes may affect hydrological systems, independent from the highly uncertain coupling of climate projections with hydrological models, which is presently the only way of assessing future hydrological changes.

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