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Geology

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Title: Sensitivity of Bunker Cave to climatic forcings highlighted through multi-annual monitoring of rain-, soil-, and dripwaters

Article Type: Research paper

Keywords: drip rate; infiltration; temperature; oxygen and hydrogen isotopes; NAO; cave microclimate monitoring

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Abstract: The last two decades have seen a considerable increase in studies using speleothems as archives of past climate variability. Caves under study are now monitored for a wide range of environmental parameters and results placed in context with speleothem data. The present study investigates trends from a seven year long monitoring of Bunker Cave, western Germany, in order to assess the hydraulic response and transfer time of meteoric water from the surface to the cave. Rain-, soil-, and dripwater were collected from August 2006 to August 2013 at a monthly to bimonthly resolution and their oxygen and hydrogen isotope composition was measured. Furthermore, drip rates were quantified. Due to different drip characteristics, annual mean values were calculated for the drip rates of each drip site. Correlations of the annual mean drip rate of each site with precipitation and infiltration demonstrate that the annual infiltration, and thus the annual precipitation control the inter-annual drip-rate variability for all except one site. The hydraulic response is not delayed. All drip sites display identical long-term trends, which suggests a draining of a common karst reservoir over these seven years of monitoring. Correlations of soil- and dripwater monthly delta180 and deltaD values with atmospheric temperature data reveal water transfer times of 3 months for soilwater at site BW 2 (40 cm depth) and 4 months for 70 cm depth (soilwater at site BW 1). Finally, the water reaches the cave chambers (15 to 30 m below land surface) after ca. 2.5 years. Consequently, a temporal offset of 29 to 31 months (ca. 2.5 years) between the hydraulic response time (no time lag on annual basis) and the water transfer time (time lag of 29 to 31 months) was found, which is negligible with regard to Bunker Cave speleothems because of their slow growth rates. Variations in drip rate and thus, precipitation and infiltration are recorded by delta13C and Mg/Ca ratios in speleothem calcite. Speleothem delta180 values reflect both temperature and precipitation signals due to drip rate-related fractionation processes. We document that long-term patterns in temperature and precipitation are

recorded in dripwater patterns of Bunker Cave and that these are linked to the North Atlantic Oscillation (NAO).

Dear Dr. Böttcher,

Please find below our response to the reviewers. We would like to emphasize that we greatly appreciate the helpful and constructive comments of the two reviewers. Their work certainly led to an improved version of our paper. In the following, detailed answers to the individual comments are provided.

Reviewer #1: Overview - This study involves an analysis of oxygen and hydrogen isotopic values in precipitation, soil water, and cave dripwater, as well as calculations of infiltration systematics and measurements of dripwater activity. The overarching goal is to better understand the response time and degree of mixing/alteration of isotopic signals between the land surface and the cave. Once these processes are constrained, speleothems can then be used most effectively to reconstruct paleoenvironmental change and thereby linked to climatic phenomena such as the NAO.

Aside from the fact that paper too often reads as a laundry list of observations, I have two major concerns. The first, as I note below, might be due to my inexperience working with vadose zone hydraulics, but involves the calculation of infiltration RATES without knowing antecedent moisture conditions and hydraulic head. Perhaps I simply missed it, but I see only calculations of permeability. I am more than happy to have my mistake pointed out to me by the authors in the revision. In any case, this section must be more carefully and fully fleshed out before it can be considered for publication. In the hydro literature, entire manuscripts are devoted to calculating infiltration rates but here it seemingly appears out of thin air next to a permeability determination. Why not just let the isotopes speak for themselves? Forget the permeability calculations and just focus on what the geochemistry of the drips vs precip tells you. The paper would be more concise and would contain less arm-waiving.

**Our reply:** We thank the reviewer for this constructive comment. The calculation of the coefficient of permeability  $(k_f)$  is a very simplified approach to calculate the transfer time through the soil. This approach ignores many important factors, which influence the infiltration of water in the soil. Therefore, we follow the suggestion of the reviewer and focus on the isotopes. As a consequence all text sections regarding the  $k_f$  value and the transfer time calculated by it are deleted.

Second, I know that there are numerous studies showing offsets between d18O values of drips collected at the same time in the same room of the same cave. Likewise, variations in d13C in drips can be ascribed to PCP, but how do you substantially alter the d18O of infiltrating water by PCP? Evaporation could do it, but the dD/d18O relationships suggest no evap is occurring.

**Our reply:** Due to statistical artifacts, we were led on the wrong path. We re-calculated all correlations with the monthly data as suggested by the second reviewer and our original conclusion that PCP alters the  $\delta^{18}$ O signal is thus invalid. We apologize for this. These parts were completely rewritten.

Line 32 - "dripwater were"

Our reply: Done (line 32).

35 - commas used incorrectly. Edit as follows: "...infiltration, and thus the annual precipitation, control ..."

Our reply: Done (line 37).

39 - what is BW2? Is this a specific sample? A location? A depth? A soil classification (B horizon...?)

**Our reply:** We clarified this part (lines 40-43) and wrote: "Correlations of soil- and dripwater monthly  $\delta^{18}$ O and  $\delta$ D values with atmospheric temperature data reveal water transfer times of 3 months for soilwater at site BW 2 (40 cm depth) and 4 months for 70 cm depth (soilwater at site BW 1). "

40 - am I interpreting this correctly? Your estimates are that water takes up to 6 months to infiltrate 40 cm into the soil?!? What sort of soil is this? If it's fine-grained, I would have thought that capillarity alone would draw water down much faster than this.

Our reply: This issue has been solved. See first main comment.

41 - this must mean that the water moves much, much faster once it enters the epikarst. How

deep is the cave below landsurface? Below soil/bedrock interface? This should be mentioned here. So far, the abstract has been a little jumpy and hard to follow.

**Our reply:** We added the information about the depth of the cave chambers below landsurface as suggested (line 43).

42 - is this a consistent offset? Is it worth mentioning the magnitude or nature of this offset here in the abstract?

Our reply: It is a consistent offset and we added the magnitude of it in the abstract (line 44).

44 - repeating the word "due" is awkward

Our reply: This sentence is deleted. See second main comment.

44 - I will be interested to read how you know that PCP is to blame; I think of PCP as influencing d13C but not necessarily O because the oxygen reservoir is so much larger than that of C.

Our reply: See second main comment.

45 - since O is affected by not H, it can't be evaporation, but I don't see (yet) how PCP can influence the d18O signal so profoundly.

Our reply: See second main comment.

49 - this abstract is a little choppy. Adding a little more information to tie the narrative together would help the reader understand the point of the study and would make for a more fluid discussion.

**Our reply:** Due to changes according to the first and second main comment and the main comments of the second reviewer, the abstract was partly rewritten.

53 - perhaps write "(for a summary, see Fairchild and Baker, 2012)"

Our reply: Done (lines 57-58).

54 - I suggest using either "Th/U" or "230Th/234U" but don't mix and match

**Our reply:** <sup>230</sup>Th/U is an established notation. See for example Scholz and Hoffmann (2008). We keep it as it is (line 59).

55 - "many, largely geochemical, parameters, " is awkwardly worded

Our reply: We replaced "many, largely" by "several, mostly" (line 60).

57 - use "major" rather than "main"

Our reply: Done (line 61).

60 - remove first use of "e.g."

Our reply: This part was rewritten and the e.g. was deleted.

73 - Do you mean to state that there have been studies on the elemental composition of rain water? I am surprised by this.

**Our reply:** We apologize for this confusing sentence. There are no studies on the elemental composition of rainwater in the study area. We rewrote the sentence to state this correctly (lines 69-72).

74 - this is the major raison d'etre for the study: to better understand cave carbonate-based paleoenvironmental reconstructions through analysis of cave hydraulics. Make that point more clear in this sentence.

**Our reply:** We rewrote the sentence as the reviewer suggested: "Furthermore, in order to link the dripwater with speleothems used for palaeoclimate reconstructions, recent cave carbonate

precipitates have been investigated in combination with their respective dripwater composition and the hydraulic regime in the cave" (lines 72-75).

79 - it may be preference only, but I suggest adding the word "values" after "d18O" or "dD".

Our reply: Done (line 80).

83 - is "whilst" the correct word here? Are you actually contrasting the Breitenbach study/interpretation with that of Riechelmann? The way it's written, it seems like both authors refer to the same cave system.

**Our reply:** We deleted "whilst" and included two sentences to clarify this matter (lines 83-86).

87 - "focused" is misspelled

Our reply: Corrected (line 88).

92 - don't use "relevant" twice in the same sentence

Our reply: We replaced the first "relevant" by "of significance" (line 93).

94 - "This paper"

Our reply: Done (line 95).

97 - "main goals of the present study"

Our reply: Done (line 98).

97 - longer than what? Be specific here. Multi-annual? Decadal?

Our reply: We added "multi-annual" after "longer-term" (line 99).

98 - what the "environmental parameters"? Temp? Precip? Be specific.

Our reply: We modified lines 95 to 96 to clarify this.

101 - delete this last sentence. It is redundant with (iv).

Our reply: Done.

103 - Here at the end of the Introduction, I think the first paragraph is a bit of a detour and suggest shortening it considerably and integrating it into the second paragraph.

**Our reply:** We shortened the first paragraph and put it together with the second one as suggested by the reviewer (lines 56-64).

106 - what does "fully humid" mean?

**Our reply:** Fully humid means there is no dry season, but equally distributed rainfall throughout the whole year. We used the Köppen-Geiger climate classification world map updated by Kottek et al. (2006). Nevertheless, we added a short explanation in brackets (lines 106-107).

109 - include 1 s.d. values of mean annual precip.

**Our reply:** Due to minor relevance of this information regarding the interpretation and discussion of our paper, we deleted the paragraph and Figure 2.

111 - include 1 s.d. values of MAAT

**Our reply:** Due to minor relevance of this information regarding the interpretation and discussion of our paper, we deleted the paragraph and Figure 2.

114 - you don't need to tell the reader it is colder in winter than in summer; delete this sentence

#### Our reply: Done.

120 - do we have any estimates of percent land surface covered by houses, roads, parking lots, sidewalks, etc? These are obviously important components of infiltration hydrology.

Our reply: The information was added as requested (lines 123-124).

123 - I am not sure "brown" is appropriate here. What about Munsell soil chart codes?123 - I would write " loamy soil formed in loess"

**Our reply:** We added the colours of the soil according to the Munsell soil chart code and defined the soil according to USDA Soil Taxonomy (lines 115-118).

126 - "... and subordinate along bedding planes" sounds awkward. I think you are missing a word in this phrase.

Our reply: We deleted "subordinate along".

138 - how was rainwater stored between (bi)monthly sample collection visits? What methods were utilized to minimize evaporation?

**Our reply:** We wrote "Rainfall amount was measured and sampled daily and collected as monthly samples between 2007 and 2012. In 2013, rainwater samples were collected only bimonthly. Rainwater samples were stored in PET bottles in a fridge to minimize evaporation." to clarify this (lines 133-136).

145 - I suggest hyphenating TS-1, etc.

**Our reply:** In order to keep the consistency with all other Bunker Cave studies, which examined the same drip sites, we prefer not to change the labelling of the sample sites.

160 - since you are converting to volumetric discharge, write the equation in the standard form as volumetric discharge (ml/time) = drip rate (drips/time) x volume/drip (ml/drips)

Our reply: Done (line 158).

165 - what is an "instantaneous water sample" and how do you know the drip volume?

**Our reply:** An instantaneous water sample is collected during the monitoring tour, therefore the exact time of collection is known. The drip volume was determined as described in the previous sentence. In order to clarify this we added this information (lines 163-164).

176 - delete hyphen

Our reply: Done (lines 175 and 178).

176 - I assume this means  $\pm 0.09\%$ ?

**Our reply:** We added "±" (lines 175 and 178).

183 - "(Fig. 1) - Hagen-Fley ... and Hemen (...) - were used..."

Our reply: Done (lines 182-184).

191 - is 13:00 a time of day?

Our reply: We replaced it by "1 p.m." (lines 190 and 195).

194 - this equation needs to be referenced to its original author

Our reply: Done (line 191).

198 - "equation was used"

Our reply: Done (line 197).

203 - ok but weather station on top of the cave would have been better

**Our reply:** The reviewer is right. The perfect solution would be a weather station directly above the cave, where precipitation amount and temperature are measured. Unfortunately, we did not have this luxury and therefore use the temperature data being from the nearest meteorological station 15 km away. Precipitation was collected at the German Cave Museum only 1.5 km from Bunker Cave (subchapter 3.1 lines 131-133).

215 - how were the soil samples collected? Please include methodology. If soils were compacted during collection, then their permeability would have been decreased.

Our reply: This issue has been solved. See first main comment.

216 - is "loam" the formal particle size distribution (e.g., silty clay loam) or a more generic term?

Our reply: This issue has been solved. See first main comment.

223 - As I recall, permeability (k) is fluid-specific, whereas hydraulic conductivity (K) is permeability specifically dealing with water. Is there a reason that k is chosen over K here?

Our reply: This issue has been solved. See first main comment.

246 - combine some of the short, choppy sentences that begin this paragraph

Our reply: Done (line 246 ff.).

251 - is the negative supposed to be in front of 171? How do you have negative infiltration? Does this mean potential ET was greater than precip by that amount? That's a very dry year in Germany.

**Our reply:** Yes, the negative is supposed to be in front of 171, see also Figure 4. 2003 was an exceptional dry year. Mean annual ET was greater than the precipitation amount and thus resulted in a negative infiltration and a dry year in this part of Germany (lines 232-233).

252 - you have already defined the acronym MAAT so use it here rather than redefining it

Our reply: Done (line 235).

253 - I find the word "whilst" to be distracting. I suggest replacing it in all case with "while"

Our reply: Done.

299 - "akin" means "similar" but isn't really correctly used in this sentence. Replace with "similar"

Our reply: This sentence has been deleted.

311 - sections 4.1 - 4.3 involve a largely mundane play-by-play of observations. Perhaps it will read better when juxtaposed to the figures on the journal page, but as it stands now, it is difficult to follow (or care about) without knowing what the point of all this is. The paper needs to include a better introduction to each section (or at least each major section) to provide a road map to the reader about what information is about to learned and why it is important and how it fits into the overall narrative of the study. For example, it is interesting and important to note that the drip sites mostly show similar trends over time, but why is this important to the study?

**Our reply:** We apologize for the problems pointed out by the reviewer. We added information regarding why these results are of importance at the beginning of the results section (lines 217-222). Furthermore, the individual sections were shortened or rewritten (see also the second main comment of the second reviewer).

316 - the abbreviation "i.e." is overused in this manuscript and is inappropriate here.

Our reply: This section was deleted. See first main comment.

320 - I am missing something here, and I recognize that it is likely rooted in my own lack of understanding of this topic, but infiltration is not solely a function of permeability. This is just one of several factors that drive infiltration. The others are the depth of ponded water (that provides the head) and the antecedent moisture (saturation state) of the soil. It is unclear to me how these infiltration rate calculations were calculated.

Our reply: This section was deleted. See first main comment.

329 - I am not exactly sure what is meant by "hydrologically connected" but at least they have similar hydraulic properties (orientation, number and size of fractures, matrix hydraulic conductivity, etc.).

**Our reply:** We rewrote the sentence to "these sites are characterized by similar hydrological properties" (line 300).

330 - drips don't respond to a "pattern", but instead they respond to some type of forcing, most likely a climatic one. Please reword.

**Our reply:** We agree with the reviewer and rewrote the sentence: "...respond to the same environmental (climate) forcing." (lines 301-302).

353 - or it might not...

**Our reply:** We agree with the reviewer. That is why we used the word "might". The correlations are possibly affected but not necessarily. We rewrote the sentence slightly to "..., which might affect the correlation" (line 326).

453 - I would like to see a citation to a study in which PCP is shown to noticeably alter d180 values. It's effects should be concentrated in the d13C value given that the carbon reservoir in infiltrating fluids is much, much smaller than the oxygen reservoir.

**Our reply:** This part has been deleted. See second main comment and first main comment of the second reviewer.

Notes on Figures.

Figures, 4, 6, and 7 - I can't distinguish between errors bars for different points (triangles vs circles) in some portions of a few panels (TS5 and BW1, as examples).

Our reply: We changed colors to better distinguish the error bars.

Reviewer #2: This is a nice study that presents the synthesis of a seven years of monitoring in Bunker Cave, NW-Germany. Even if it is already published, I think that a complete figure with all the monthly data is necessary here so that we can appreciate the seasonality of the rainfall d180 and the more or less stability of the dripping cave water. This cave is a specific site that is covered by a thick soil which slows down the rainfall infiltration but this is not the main reason and the important result in this study is certainly the quite long time (up to 3 years) that meteoric water takes to reach the cave galleries. This is an important result that highlights the importance of a long monitoring in order to understand the infiltration processes and for this reason it merits publication.

However there are two major points that needs more explanations and/or different interpretations:

1) I am not convinced by the fact that prior calcite precipitation (PCP) changes the d180 composition of seepage water because the molecule quantity of water containing oxygen is much more important than HCO3- ions that are involved in the calcite precipitation. Moreover, that fact that there is no difference between instantaneous and monthly collected water is in contradiction of a PCP influence on the d180. PCP which is likely seasonal in most sites, would then produce differences between summer and winter dripping waters (?). I suggest that the authors check this possibility by measuring Mg/Ca and Sr/Ca and doing Ca-Mg/Ca and Ca-Sr/Ca graphs. This is an important point on which all the work that is (and was) done on fluid inclusions depends;

**Our reply:** Statistical artifacts led to a misinterpretation of the data. We apologize for that. Thus, the original interpretation that PCP alters  $\delta^{18}$ O is invalid. The respecting passages have been deleted and rewritten. See also second main comment by the first reviewer. Furthermore, we followed the suggestion by the reviewer and present the monthly data of  $\delta^{18}$ O and  $\delta$ D of rain-, soil- and dripwater. 2) In the results part, you have to explain clearly, in the main text, how you calculate the d18O annual means : what limits did you take , month of beginning and month of end . Please explain how you choose these limits. Also, you have to precise the months during which the infiltration occurs (winter and spring? part of Fall or most of the year). But, the way used to calculate the mean d18O of rainwater is already an interpretation because it is calculated on ONE year. It could be actually much more or less; you may find this by doing weighted averaging from month to month (in the past) until reaching the d18O value of the dripping site. Finally, considering the surface air temperature as a proxy of the d18O seems hazardous unless you have strong correlations between T and rainwater d18O.

**Our reply:** At the beginning of the results section we added information on the calculation of the annual mean values. Furthermore, we added the time of main infiltration in lines 233-234. We followed the reviewer's suggestion to use the monthly values to calculate the water transfer time. However, since the GNIP stations are far away from the study site and  $\delta^{18}$ O and  $\delta$ D values of rain water of the monitoring only cover the monitoring period, we used the atmospheric temperature of the close-by meteorological stations. The reviewer is correct by pointing out that this approach is only feasible, when  $\delta^{18}$ O and  $\delta$ D of rainwater show a strong correlation with T. Since this is the case as we pointed out in section 4.1 lines 253-256 (citing Riechelmann et al., 2011) and section 5.2 lines 368-369, we consider this approach as valid. According to this new approach results (4.3) and interpretation and discussion (5.2) sections were rewritten.

#### Detailed comments:

About references: when speaking about generalities, I think that it would be better to refer to precise studies that marked the topics. For example, line 56 and elsewhere, instead of referring to the nice book of Fairchild and Baker 2012, it would be better to refer to Cheng et al. or Hellstrom et al. or other specialists in U-Th that really worked on the topics; the same for other subjects; otherwise the Fairchild and Baker book will be referred in all cases.

Our reply: We replaced the Fairchild and Baker citation by other studies.

Lines 58-59 : what drove this choice of references ? If it is a globally important like results on speleothem stable isotopes, you may cite Cheng et al. 2016 Nature which presents an

unprecedented well dated 600ka speleothem record; you may also focus on more regional studies.

**Our reply:** The choice of references should be representative for the region. Therefore, we cite original studies from western central Europe. We feel that the Cheng et al. (2016) study, being focused on the Asian monsoon, is not appropriate here (lines 62-63).

Lines 90-91: I do not entirely agree that nobody before detrended the seasonal signal. In their work on Villars and Chauvet caves, Genty et al. (GCA 2014) made rainfall averaging on several years (up to > 10 yrs) which is a real long term analysis.

**Our reply:** We corrected this (line 91).

Lines 101-102 : not necessary in an article

Our reply: This sentence was deleted.

Lines 153-155 : don't you think that the fact that there is no difference between instantaneous and monthly collected water is in contradiction of a PCP influence on the d18O ? PCP which is likely seasonal in most sites, would then produce differences between summer and winter dripping waters (?).

**Our reply:** The reviewer is right. PCP does not alter  $\delta^{18}$ O in Bunker Cave. See also first main comment.

Line 194 : dealing with temperatures, I guess that it is 273 instead 237 (?)

**Our reply:** This value is correct (237; line 193).

Lines 293-295 : this sentence is not clear to me

Our reply: The sentence was rewritten (lines 282-283).

Lines 351-353 : I entirely agree with this. Do you have any idea of the duration, in the year, of

the real infiltration ? Or, what are the months, in the year, during which the rainfall infiltrate ? You may find this by doing weighted averaging from month to month (in the past) until reaching the d18O of the dripping site.

Our reply: We added the information of infiltration time in this paragraph (lines 324-326).

In summary, these were constructive reviews, which provided helpful suggestions for improvement of this manuscript that we submit as a revised version.

Sincerely

Sylvia Riechelmann on behalf of the authors.

1	Sensitivity of Bunker Cave to climatic forcings highlighted through multi-annual
2	monitoring of rain-, soil-, and dripwaters
3	
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19	
20	Chemical Geology
21	
22	Keywords: drip rate, infiltration, temperature, oxygen and hydrogen isotopes, NAO, cave
23	microclimate monitoring
24	
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1

#### 26 ABSTRACT

The last two decades have seen a considerable increase in studies using speleothems as 27 archives of past climate variability. Caves under study are now monitored for a wide range of 28 environmental parameters and results placed in context with speleothem data. The present 29 study investigates trends from a seven year long monitoring of Bunker Cave, western 30 Germany, in order to assess the hydraulic response and transfer time of meteoric water from 31 the surface to the cave. Rain-, soil-, and dripwater were collected from August 2006 to August 32 2013 at a monthly to bimonthly resolution and their oxygen and hydrogen isotope 33 composition was measured. Furthermore, drip rates were quantified. Due to different drip 34 characteristics, annual mean values were calculated for the drip rates of each drip site. 35 Correlations of the annual mean drip rate of each site with precipitation and infiltration 36 demonstrate that the annual infiltration, and thus the annual precipitation control the inter-37 38 annual drip-rate variability for all except one site. The hydraulic response is not delayed. All drip sites display identical long-term trends, which suggests a draining of a common karst 39 reservoir over these seven years of monitoring. Correlations of soil- and dripwater monthly 40  $\delta^{18}$ O and  $\delta$ D values with atmospheric temperature data reveal water transfer times of 3 41 months for soilwater at site BW 2 (40 cm depth) and 4 months for 70 cm depth (soilwater at 42 site BW 1). Finally, the water reaches the cave chambers (15 to 30 m below land surface) 43 after ca. 2.5 years. Consequently, a temporal offset of 29 to 31 months (ca. 2.5 years) between 44 the hydraulic response time (no time lag on annual basis) and the water transfer time (time lag 45 of 29 to 31 months) was found, which is negligible with regard to Bunker Cave speleothems 46 because of their slow growth rates. Variations in drip rate and thus, precipitation and 47 infiltration are recorded by  $\delta^{13}$ C and Mg/Ca ratios in speleothem calcite. Speleothem  $\delta^{18}$ O 48 values reflect both temperature and precipitation signals due to drip rate-related fractionation 49 processes. We document that long-term patterns in temperature and precipitation are recorded 50

in dripwater patterns of Bunker Cave and that these are linked to the North AtlanticOscillation (NAO).

53

#### 54 **1. Introduction**

55

Palaeoclimate reconstructions based on speleothems, i.e. mostly carbonate deposits 56 formed in caves, have increased significantly during the last two decades (for a summary, see 57 Fairchild and Baker, 2012). The most important strengths of speleothems are the precise 58 <sup>230</sup>Th/U dating (e.g., Dorale et al., 2004; Scholz and Hoffmann, 2008; Cheng et al., 2013) and 59 the availability of several, mostly geochemical, parameters such as carbon and oxygen isotope 60 values, major and trace elemental abundances, and  $\delta^{18}O$  and  $\delta D$  of fluid inclusions (e.g., 61 Niggemann et al., 2003; Mangini et al., 2005; Vonhof et al., 2006; Fohlmeister et al., 2012; 62 Scholz et al., 2012; Luetscher et al., 2015). These can be used for single- or multi-proxy 63 approaches to reconstruct past climate dynamics. 64

In order to gain a better understanding of the processes influencing geochemical 65 proxies in the soil and epikarst zone, as well as processes acting during deposition of 66 speleothems, sophisticated monitoring programmes have been established (e.g., Spötl et al., 67 2005; Mattey et al., 2008b, 2016; Riechelmann et al., 2011, Wassenburg et al., 2013; Genty et 68 al., 2014; van Rampelbergh et al., 2014, Breitenbach et al., 2015; Treble et al., 2016). In the 69 context of these efforts, cave air temperature, pCO<sub>2</sub> and humidity, drip rate, as well as the 70 isotopic composition of rain-, soil- and dripwater and the element concentrations of soil-, and 71 dripwater have been recorded. Furthermore, in order to link the dripwater with speleothems 72 used for palaeoclimate reconstructions, recent cave carbonate precipitates have been 73 investigated in combination with their respective dripwater composition and the hydraulic 74

regime in the cave (e.g., Miorandi et al., 2010; Tremaine et al., 2011; Riechelmann et al.,
2013, 2014).

Most monitoring studies focused on seasonal variations of the above-mentioned 77 parameters, whereby, for example, the drip rate was analysed to study the response to rainfall 78 events or the hydrological connection of the drip sites (Baker et al., 1997; Mattey et al., 79 2008b; Riechelmann et al., 2011). The analysis of dripwater  $\delta^{18}$ O and  $\delta$ D values shows either 80 81 seasonal variations (Mattey et al., 2008a, Breitenbach et al., 2015) or rather stable values close to the annual mean of rainwater (Riechelmann et al., 2011; van Rampelbergh et al., 82 83 2014). Seasonal variations in cave dripwater reflect a fast transfer of the water (e.g., Breitenbach et al., 2015). The lack of an intra-annual pattern points to strong mixing in the 84 epikarst and/or the vadose zone with transfer times between the soil and the cave drip site in 85 excess of one year (e.g., Riechelmann et al., 2011; van Rampelbergh et al., 2014). Financial 86 constraints, accessibility, or lack in manpower commonly limit monitoring studies to 87 durations of a few years or less. Thus far, only a very limited number of studies focused on 88 multi-annual trends of the monitored parameters and implications for speleothem research 89 (Genty and Deflandre, 1998; Treble et al., 2013; Genty et al., 2014, Breitenbach et al., 2015; 90 Mattey et al., 2016). To the knowledge of the authors, only the study by Genty et al. (2014) 91 92 detrended the seasonal signal in order to gain insights in potential longer-term (> 5yrs) trends. This is of significance as particularly the longer-term trends are relevant for the assessment of 93 proxy data from speleothems recording decadal or longer variability only. 94

This paper documents and discusses observations of precipitation/infiltration and drip rate as well as atmospheric temperature and the oxygen and hydrogen isotopic composition of rain-, soil-, and cave dripwater from a seven year-long monitoring campaign in Bunker Cave in northwestern Germany. The main goals of the present study are: (i) to quantify the longerterm (multi-annual) variability of the environmental parameters; (ii) to assess the response time of the carbonate precipitating drip sites to these parameters; (iii) to identify signal
smoothing and possible alteration processes during percolation through the epikarst; and (iv)
to draw, where possible, general implications for speleothem research.

103

#### 104 2. Climate and cave parameters

105

The climate of northwestern Germany is warm-temperate, i.e. fully humid (equally distributed rainfall amount throughout the year) with warm summers (Kottek et al., 2006). Bunker Cave is located between the villages of Iserlohn and Letmathe in the Rhenish Slate Mountains in the NW part of the Sauerland, Germany (Fig. 1). It is part of the Bunker-Emst-Cave system (3.5 km long) with the Bunker Cave entrance (51°22'N, 07°40'E) being located at 184 m above sea level (asl) on a south-facing hill slope of the Dröscheder Emst karst plateau (Grebe, 1993; Hammerschmidt et al., 1995).

The host rock consists of Middle to Upper Devonian massive limestone (von Kamp, 113 1972). The rock overburden of the cave is between 15 and 30 m (Grebe, 1993) and the host 114 rock is overlain by ca. 70 cm inceptisol to alfisol (USDA Soil Taxonomy). The colour varies 115 between dark and yellowish brown (10YR 3/3 and 10YR 5/6) for the upper soil layers and 116 bright reddish brown to bright brown (5YR 5/8 to 7.5YR 5/8) for the lower layer (Munsell 117 soil colour charts). The vegetation above the cave consists of deciduous forest (mainly ash 118 and beech trees) and scrubs (Riechelmann et al., 2011). Bedding dips to the North or 119 Northwest (von Kamp and Ribbert, 2005) and water percolates mainly along fractures and 120 bedding planes. This feature and the fact that Bunker Cave is located in a south-facing hill 121 reduces the effective catchment of the cave dripwater to a few hundred  $m^2$ . Furthermore, the 122 Dröscheder Emst karst plateau is partly used as a residential area and ca. 15 to 20% of the 123 catchment area is anthropogenically sealed. Furthermore, a railway route runs above the cave. 124

125

#### 126 **3. Materials and Methods**

127

#### 128 3.1 Monitoring and sample collection of rain-, soil- and dripwater

129

The monitoring programme performed in and above Bunker Cave ran from August 2006 to August 2013. Rainwater samples were collected with a rain gauge according to the DIN 58666C norm on the roof of the German Cave Museum Iserlohn (51°22'N, 07°38'E; 175 m asl), located 1.5 km from Bunker Cave (Fig. 1). Rainfall amount was measured and sampled daily and collected as monthly samples between 2007 and 2012. In 2013, rainwater samples were collected only bimonthly. Rainwater samples were stored in PET bottles in a fridge to minimize evaporation.

Two soilwater sampling sites were installed above Bunker Cave. The soilwater suction probes were manufactured by Umwelt-Geräte-Technik GmbH (UGT, Germany). One probe (BW 1) was installed at a depth of 70 cm and water was sampled monthly between 2007 and 2011. The second probe (BW 2) sampled water at 40 cm depth in monthly intervals between 2009 and 2011. In 2012 and 2013, both soilwater sites were sampled only bimonthly.

In total, five drip sites (TS 1, TS 2, TS 3, TS 5 and TS 8; Fig. 2) were monitored in 142 Bunker Cave from 2006 to 2013. Sampling at drip site TS 8 started in 2007. Drip sites TS 1 143 and TS 5 are located in chamber 1. All other sites are located in chamber 2 of Bunker Cave 144 (Fig. 2). Dripwater samples were integrated over one month in order to obtain sufficient 145 volumes of water for multi-proxy geochemical analyses. These monthly samples were taken 146 between 2006 and 2011, while bimonthly samples were taken between 2012 and 2013. An 147 exception is drip site TS 1, where sufficient water could be collected during each visit. 148 Dripwater first dripped onto a drip counter placed in a plastic box and was then transferred 149

150 into a closed bottle via tubing to minimize  $CO_2$  degassing and potential evaporation. 151 Comparison of instantaneous and monthly collected water revealed similar  $\delta^{18}O$  values, 152 confirming that fractionation processes due to evaporation are negligible (Riechelmann et al., 153 2011). For further details of the monitoring, the reader is referred to Riechelmann (2010) and 154 Riechelmann et al. (2011; 2012a; 2013; 2014).

Drip rates were measured manually with a stopwatch. The following equation was used to convert drip rate into volumetric discharge (*z*):

157

158  $z \,[\text{ml/time}] = \text{drip rate [drips/time]} * V \,[\text{ml/drips}]$  (1)

159

where V is the volume of each drip. The volume of a drip was determined by a defined 160 collection time, the total amount of water and the drip rate (in drips per min). In some cases, 161 the drip rate at site TS 8 was not quantified by means of a stopwatch, and was instead 162 calculated via the amount of an instantaneous water sample (collected during the monitoring 163 tour) and the known drip volume (determined as described above). Furthermore, drip rates 164 were measured automatically by acoustic drip counters (Stalagmate; Mattey and Collister, 165 2008), which were placed under drip sites TS 2 (2009-2013), TS 3 (2010-2013), TS 5 (2007-166 2013) and TS 8 (2009-2013). 167

168

### 169 3.2 $\delta^{l8}O$ and $\delta D$ of rain-, soil- and dripwater

170

The oxygen and hydrogen isotopic compositions of all water samples were determined at the University of Innsbruck. Water samples were collected in small, airtight glass vials without headspace. The oxygen isotope composition was analysed using a ThermoFinnigan DELTA<sup>plus</sup>XL mass spectrometer connected with a Gasbench II using the CO<sub>2</sub> equilibrium technique. The 1σ reproducibility of the  $\delta^{18}$ O values is ±0.09 ‰ VSMOW (Spötl et al., 2005). The hydrogen isotope composition was measured using a ThermoFinnigan DELTA<sup>plus</sup>Advantage equipped with a TC/EA high-temperature pyrolysis unit. The 1σ reproducibility is ±1 ‰ VSMOW.

179

#### 180 *3.3 Evapotranspiration and infiltration*

181

Instrumental climate data of two nearby meteorological stations (Fig. 1) - Hagen-Fley
(51°25'N, 07°29'E; 100 m asl; 2000-2007; www.dwd.de) and Hemer (51°23'N, 07°45'E; 200
m asl; 2008-2013; www.meteomedia.de) - were used to calculate the evapotranspiration after
Haude (1955) using the following equation:

186

187 
$$E_{pot} [\text{mm/day}] = x [\text{mm/day} * \text{hPa}] * P^{13} [\text{hPa}] * (1 - (F^{13} [\%] / 100))$$
 (2)

188

189 where  $E_{pot}$  is the potential evaporation, *x* represents the monthly coefficient depending on 190 vegetation,  $P^{13}$  is the saturation pressure at 1 p.m. and  $F^{13}$  is the relative humidity at 1 p.m.. 191 The saturation pressure was calculated using the Magnus-formula:

192

193 
$$P^{I3}$$
 [hPa] = 6.107 [hPa] \*10^((7.5 \*  $T^{I3}$  [°C]) / (237 +  $T^{I3}$  [°C])) (3)

194

195  $T^{l3}$  is the air temperature at 1 p.m.. Equations 2 and 3 were used to calculate the amount of 196 water which can potentially infiltrate ( $Inf_{pot}$ ) into the soil and thus, into the cave. Therefore, 197 the following equation was used:

198

199 
$$Inf_{pot} [mm/day] = N [mm/day] - E_{pot} [mm/day]$$
 (4)

200

where *N* is precipitation. The amount of water infiltrating into the soil above Bunker Cavewas determined using precipitation data from the German Cave Museum Iserlohn.

203

204 *3.4 GNIP stations* 

205

Data of four nearby GNIP (Global Network of Isotopes in Precipitation; http://wwwnaweb.iaea.org/napc/ih/IHS\_resources\_gnip.html) stations were used for comparison with monitoring and meteorological data. These include Bad Salzuflen (52°06'N, 08°43'E; 100 m asl), Emmerich (51°49'N, 06°36'E; 43 m asl), Koblenz (50°21'N, 07°34'E; 97 m asl) and Wasserkuppe Rhön (50°30'N, 09° 57'E; 921 m asl).

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212 4. Results
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213

Seasonally resolved monitoring data of rain-, soil- and dripwater of Bunker Cave were 214 partly published by Immenhauser et al. (2010), Kluge et al. (2010, 2013), Riechelmann et al. 215 (2011, 2012a, b, 2013, 2014), Fohlmeister et al. (2012), Münsterer et al. (2012), and 216 Wackerbarth et al. (2012). In this study, we present monthly data as well as annual mean 217 values of the relevant parameters, which were calculated using the monthly or bimonthly 218 values of the respective year to (i) examine long-term variability and (ii) compare these data 219 with precipitation/infiltration and temperature data to determine the hydraulic response time 220 and water transfer time. Annual means were obtained from monthly data (January to 221 December). 222

223

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224 4.1 Precipitation, infiltration, temperature and rainwater \delta^{18}O and \delta D
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226 For this study, the time period from 2000 to 2013 is considered. Annual mean values are shown for all years except 2013, since monitoring stopped in August of this year. Hence, 227 all mean values for 2013 are related to this time interval. Furthermore, annual mean values for 228 2006 were not calculated, due to insufficient data. The lowest annual precipitation sum (720 229 mm) was recorded in 2003, without considering 2013 with the lowest precipitation sum of 230 231 447 mm. The highest annual rainfall (1202 mm) and the highest mean annual infiltration (546 mm) occurred in 2007 (Fig. 3A). The lowest annual mean infiltration was observed in 2003 (-232 171 mm). Infiltration at Bunker Cave is lower during summer months than during winter 233 (Riechelmann et al., 2011). Monthly temperature data show low values in winter and high 234 ones during summer. The lowest mean annual air temperature (7.1°C) was observed in 2010, 235 while the highest (11.3°C) was observed twice, in 2000 and 2007 (Fig. 3A). 236

Oxygen and hydrogen isotope values of monthly rainwater samples display a seasonal variability with higher values during summer and lower ones during winter months (Fig. 3B). In the years 2010 and 2013, the lowest mean annual  $\delta^{18}$ O and  $\delta$ D values of rainwater were observed ( $\delta^{18}$ O: -9.20 and -9.01‰;  $\delta$ D: -66.2 and -62.5‰), while the highest mean annual values were observed in 2011 ( $\delta^{18}$ O: -5.92‰;  $\delta$ D: -45.8‰; Fig. 3B). Monthly  $\delta^{18}$ O and  $\delta$ D values of rainwater are highly correlated (r = 0.98, p < 0.001; n = 65; see also Fig. 4C).

Since monthly rainwater samples collected during the monitoring do not cover the same time period as the monthly samples from the GNIP stations, mean annual values are used here for comparison. Mean annual oxygen and hydrogen isotope values of rainwater collected at the German Cave Museum correlate well with those of nearby GNIP stations (Figs. 4A, B). The Local Meteoric Water Line (LMWL) of the rainwater (Fig. 4C) is close to that of the GNIP station Bad Salzuflen ( $\delta D = 7.72 * \delta^{18}O + 5.7$ ; r = 0.98; Stumpp et al., 2014). Thus, rainwater collected during the monitoring period reflects the climate conditions of western Germany. The slope of 7.72 is close to that of the Global Meteoric Water Line (8), indicating insignificant evaporation of the collected samples (Gat et al., 2000). Since all dripwater data plot on the LMWL (Fig. 4C) evaporation in the soil, epikarst and cave generates no significant imprint on their corresponding isotope values. Furthermore, both isotope systems depend on the regional surface air temperature and the isotopic composition of Bunker Cave dripwater reflects the infiltration weighted mean annual  $\delta^{18}$ O and  $\delta$ D values of rainwater (Riechelmann et al., 2011).

257

258 *4.2 Drip rate* 

259

Due to different drip characteristics of the different drip sites (ranging from seepage 260 flow to seasonal drip; Riechelmann et al., 2011; Appendix S1 and Fig. 8), annual mean drip 261 rates were calculated and their long-term variability compared. Both manually and 262 automatically obtained mean annual drip rates show the same trends (Fig. 5). Sites TS 1, TS 2 263 and TS 5 show a decreasing trend in mean drip rates over the monitoring period, which is also 264 reflected in cross-correlations of mean annual drip rates at these sites (TS 1 vs. TS 2: r = 0.82; 265 TS 1 vs. TS 5: r = 0.85; TS 2 vs. TS 5: r = 0.88;  $p_{all} \le 0.02$ ,  $n_{all} = 7$ ). The annual mean drip 266 rate at site TS 3 correlates only with the annual mean drip rate at site TS 2 within the 95% 267 confidence level (r = 0.81) and with TS 1 (r = 0.79) and TS 5 (r = 0.72) just below the 95% 268 confidence level  $(n_{all} = 6)$ . However, it can be assumed that the annual mean drip rate at site 269 TS 3 follows the same trend as sites TS 1, 2 and 5. At drip site TS 8 a similar annual mean 270 drip rate was observed in 2008 and 2009 before drip rates increased until 2011, followed by a 271 decreasing trend. Drip rate of drip site TS 8 does not show any significant correlation with 272 any other drip rate. 273

274

276

Seasonal trends are difficult to identify for soilwater, because of lack of data (no 277 water), especially summer months. An exception is soilwater at site BW 1 which yielded a 278 complete time series from 2007 to 2008. There, higher  $\delta^{18}O$  and  $\delta D$  values occur during 279 summer/autumn months, while lower values are observed in winter/spring months (Fig. 6). 280 The lowest mean annual  $\delta^{18}$ O and  $\delta$ D values occurred in 2010 and 2011 for both soilwaters, 281 while the highest values are shown in 2007 and 2008 (Fig. 6). Monthly  $\delta^{18}O_{BW1}$  values are 282 significantly correlated with  $\delta^{18}O_{BW2}$  values (Table 1). The same can be observed for  $\delta D$ 283 (Table 2). 284

Seasonal trends in  $\delta^{18}$ O and  $\delta$ D of dripwaters are minor, but most pronounced for 285 water at site TS 1. In general, lower isotope values occur in summer, while higher values are 286 observed in winter (Fig. 6). Correlations between  $\delta^{18}$ O and  $\delta$ D values of monthly samples of 287 the same site are significant for all dripwaters and both soilwater samples (r > 0.44; p < 0.001; 288  $n \ge 21$ ). All dripwaters share similarities in their oxygen and hydrogen isotopic composition 289 as shown by positive cross-correlations (Tables 1 and 2). Mean annual values display the 290 overall trend for  $\delta^{18}$ O and  $\delta$ D, which is similar for all drip sites considering the errors (Fig. 6). 291 Oxygen and hydrogen isotope values increased until 2009, reached a plateau and decreased 292 around 2012. This trend is less distinct for dripwater at site TS 8 (Fig. 6). 293

294

#### 295 **5. Interpretation and Discussion**

296

297 *5.1 Constraints on the hydraulic response time* 

298

The positive correlations of mean annual drip rates of all sites except TS 8 (Fig. 5) 299 suggests that: (i) these sites are characterized by similar hydrological properties (Baker et al., 300 1997), and (ii) the drip rates of these sites most likely respond to the same environmental 301 302 (climate) forcing. According to Riechelmann et al. (2011), no drip site showed a direct response to rainfall events, but rather a lagged response of up to several months due to slowly 303 increasing hydrological pressure in the karst reservoir above the cave. Correlations between 304 305 drip rates and precipitation and infiltration during the preceding years based on monitoring 306 and meteorological data of the stations Hagen-Fley and Hemer were used to determine the response time of mean annual drip rates to mean annual precipitation and infiltration. The 307 308 correlation coefficients (r) between drip rate and infiltration show that the mean annual drip rates at TS 1 and TS 3 are controlled by mean annual infiltration of the same year, as observed 309 for other cave systems (Fig. 7A; Genty and Deflandre, 1998; Baker et al., 2000; Miorandi et 310 311 al., 2010; Tremaine and Froelich, 2013). Correlation coefficients at drip sites TS 2 and 5 are close to the 95% confidence level. Since the infiltration amount is controlled by precipitation 312 313 amount and temperature (water loss due to evapotranspiration), correlations between mean 314 annual drip rate and annual precipitation amount were calculated (Fig. 7B). Positive correlations at the 95% confidence level were found for TS 2 and TS 3 for the same year 315 interval, while correlation coefficients of TS 1 and 5 are just below the 95% confidence level. 316 Several sources of error must be considered, however, which lead to a lack of correlation at 317 the 95% confidence level. Lack of data (< 12 for annual means) may be the problem in case of 318 drip site TS 5, since neither infiltration nor precipitation correlate at the 95% confidence level 319 with drip rate. This might also be the case for sites TS 1 and 2. The calculated mean annual 320 drip rate of the former might be biased, since it shows pronounced seasonal drip rate 321 variability, with most changes not being covered by the monitoring (Figs. 5 and S1 in 322 supplementary material). Finally, the calculated mean annual infiltration in the soil reflects 323

the *potential* rather than *effective* infiltration. Furthermore, it must be kept in mind that effective infiltration is biased towards winter (Wackerbarth et al., 2010; Riechelmann et al., 2011), which might affect the correlation.

A decrease of the drip rate was already observed for two other drip sites and TS 2 in Bunker Cave for the first three years as reported in Riechelmann et al. (2011). As shown here, this trend continued (Fig. 5), i.e. the reservoir was drained further during the seven years. This might be due to relatively dry years with low infiltration at the beginning of the century. The following years (2007 to 2010) with higher infiltration most likely did not fill the reservoir sufficiently to stop the draining and afterwards infiltration decreased again (2011 to 2013; Fig. 3A).

The mean annual drip rate at site TS 8 shows a significant positive correlation with mean annual infiltration with a lag of three years, but no significant correlation near the 95% confidence level with precipitation. Drip site TS 8 is the only site, whose drip rate does not show the same trends as the other sites and also lacks a correlation with infiltration and precipitation for the interval recorded by the other drip sites. This absence of any correlation might be due to a change in drip behaviour during the monitoring period.

The drip characteristics of all drip sites, plotted according to Smart and Friederich 340 (1987) and Baker et al. (1997; see Figs. 8 and S1 in supplementary material to this paper) 341 highlight minor variability, with more or less the same drip characteristics over time. Drip 342 behaviour changed drastically at TS 8 during the monitoring period (Fig. 8): while seasonal 343 drip characteristics prevailed in 2008 and 2009, this drip shows seepage flow behaviour 344 between 2010 and 2012. Possible reasons for this change in drip characteristics include: (i) the 345 flow paths and the reservoir behaviour in the epikarst underwent changes as a result of 346 strongly variable rainfall events (Tooth and Fairchild, 2003), (ii) the flow paths of percolating 347 water changed or became blocked due to prior calcite precipitation (PCP; Fairchild et al., 348

2006) occluding the flow paths of water; (iii) the overhanging stalactite was broken off 349 (earthquake, human interference etc.) resulting in drip parameter changes (Baldini et al., 350 2006). Amongst these scenarios, it is conceivable that rainfall filled the reservoir feeding TS 8 351 to a threshold resulting in a more continuous water flow into the cave. PCP is a more likely 352 explanation for the change in drip behaviour as documented for this site (Riechelmann et al., 353 2011). The stalactite was removed in June 2008, but the change in drip rate lagged by one 354 year, suggesting that the drip pattern was not controlled by the stalactite and thus the third 355 option mentioned above is considered unlikely. Concluding, the drip rate at site TS 8 is 356 controlled by infiltration and hence rainfall, but the dripwater signal reflecting both of these 357 parameters is strongly affected by prior calcite precipitation in the aquifer. Although the 358 catchment of the cave is partly sealed, infiltration and thus the precipitation signal can still be 359 recognized in the drip rate. 360

361

#### 362 5.2 Constraints on the water transfer time

363

Water transfer times from the surface to the cave are highly variable and range from 364 days to several years not only for different caves but also for different drip sites in a given 365 cave (e.g., Kaufmann et al., 2003; Mattey et al., 2008b, Lambert and Aharon, 2010; Treble et 366 al., 2013; Genty et al., 2014, Breitenbach et al., 2015). Here, the dependency of rainwater 367  $\delta^{18}$ O and  $\delta$ D on temperature ( $\delta^{18}$ O versus T: r = 0.73;  $\delta$ D versus T: r = 0.68;  $n_{both} = 67$ ;  $p_{both} < 100$ 368 0.001; Riechelmann et al., 2011) is used to quantify the water transfer time. Correlation 369 coefficients (r) between  $\delta^{18}$ O and  $\delta$ D of soil- and dripwater with surface air temperature were 370 calculated using data of the meteorological stations Hagen-Fley and Hemer. To this end, 371 monitoring data were correlated with temperature data of preceding years. Since rainwater 372

373  $\delta^{18}$ O and  $\delta$ D values correlate positively with temperature, only positive correlations are 374 relevant to determine the water transfer time.

Monthly  $\delta^{18}$ O and  $\delta$ D values of soilwater site BW 1 at 70 cm correlate significantly 375 with the atmospheric temperature with a lag of four months (Fig. 9). In case of soilwater site 376 BW 2 a significant correlation was found using a three months lag (40 cm; Fig. 9). However, 377 gaps in the monthly soilwater data during dry summer months limit the significance of these 378 calculations and they should therefore be treated with caution. Thus, this is also resulting in 379 other highly significant correlations at 63 months lag time in case of BW 2. Since  $\delta^{18}O$  and 380  $\delta D$  values of soilwater correlate significantly with atmospheric temperature at both sites, and 381 monthly  $\delta^{18}$ O and  $\delta$ D values also correlate for both soilwater sites, we conclude that water 382 isotopes are not or only slightly altered by processes in the soil zone such as evaporation and 383 uptake of biogenic  $CO_2$  into the water (e.g., Lachniet, 2009). Thus, the temperature 384 dependency of rainwater is preserved in the  $\delta^{18}O$  and  $\delta D$  values of soilwater above Bunker 385 Cave. 386

Because dripwater at site TS 1 was collected as an instantaneous sample, the  $\delta^{18}$ O and 387  $\delta D$  values are not representative for the whole month as is the case for the other dripwaters. 388 Therefore, calculations with monthly data were only performed for all other drip sites 389 resulting in a lag of 29 to 31 months (Fig. 10). Difficulties occurred for TS 2, TS 3, and TS 8 390 in case of correlations using  $\delta^{18}$ O. There, the highest correlations are found between 41 and 391 66 months and only slightly lower ones are observed for a lag of 29 to 31 months. 392 Calculations using  $\delta D$  of all drip sites except TS 2 and  $\delta^{18}O$  for TS 5 do not show these 393 features, which are most likely due to different mixing of the younger and older water in the 394 reservoir as a consequence of different transfer times. Variability in the transfer times 395 between different drip sites may arise due to: (i) variable thickness of the soil layer above 396

Bunker Cave, and (ii) differential thickness of the host rock overlying the cave, as well as (iii) 397 differential lengths and types of individual flow paths (e.g., Tooth and Fairchild, 2003). The 398 water retention capacity of soil is another factor that may or may not lead to an admixture 399 waters with different residence time in the soil and epikarst zone (Hölting and Coldewey, 400 2013). Following dry periods, the portion of older water in the soil increases (Kottek et al., 401 2006) and the opposite is found during more humid periods. Although, rainfall is equally 402 distributed throughout the year, infiltration is not, as it is lower during summer than during 403 winter months (Wackerbarth et al., 2010; Riechelmann et al., 2011). Thus, water retention 404 capacity might have a small effect on water transfer times at Bunker Cave sulphate and nitrate 405 data show that dripwaters at sites TS 1 and TS 5 have shorter residence times compared to 406 sites TS 2, 3, and 8 (Riechelmann et al., 2011). This might explain why both  $\delta^{18}O$  and  $\delta D$ 407 correlations with temperature performed well for TS 5. Drip water at TS 2 appears to be the 408 site with the longest and strongest buffering in the epikarst zone. In contrast to most others 409 this site displays seepage flow characteristics (see Appendix S1) and the oldest age based on 410 tritium data (Riechelmann et al., 2012a). This pattern might explain why the correlations at 411 this site are weakest. However, the shift in correlations using  $\delta D$  compared to the other drip 412 sites (Fig. 10) remains insufficiently explained. Since  $\delta^{18}$ O and  $\delta$ D values of dripwater at site 413 TS 1 correlate with those of all other drip sites it can be assumed that the TS 1 water has a 414 similar transfer time of 29 to 31 months. 415

The results shown here suggest that the dripwater transfer time at all monitored drip sites in Bunker Cave ranges between 29 to 31 months (ca. 2.5 years). This is in good agreement with tritium data, suggesting a transfer time of 2-4 years (±1 year; Kluge et al., 2010). The tritium-based estimates, however, represent mixing ages of younger infiltrating water and older water of the epikarst reservoir (Kluge et al., 2010; Riechelmann et al., 2012a). 421 Based on the fact that all dripwaters plot on the LMWL, alteration of  $\delta^{18}$ O and  $\delta$ D values are 422 excluded and hence, both retain the temperature signal of rainwater.

Summing up, the multi-annual monitoring programme at Bunker Cave sheds light on
the complex processes affecting water transfer times. Obviously, a short-term monitoring
program would fail to resolve these patterns.

426

427 5.3 Influence of the North Atlantic Oscillation on the climate in western Germany

428

429 The North Atlantic Oscillation (NAO) regulates Northern Hemisphere climate variability and particularly so in western Europe and eastern North America (e.g., Marshall et 430 al., 2001; Visbeck et al., 2001; Baldini et al., 2008; Hurrell and Deser, 2010; Langebroek et 431 al., 2011; Pinto and Raible, 2012; Trouet et al., 2012; Wassenburg et al., 2013, 2016; Comas-432 Bru et al., 2013). The NAO index is defined as the difference of sea-level pressure between 433 434 the Icelandic Low and the Azores High (Hurrell, 1995; Wanner et al., 2001). While the sealevel pressure pattern is present throughout the year, it is most pronounced during winter 435 (Wanner et al., 2001; Pinto and Raible, 2012). Thus, most often the winter (December to 436 March) NAO index is used when studying climatic variability in the North Atlantic realm 437 (Proctor et al., 2000; Trouet et al., 2009; Baker et al., 2015). During a negative NAO mode, 438 the winter in Europe is dominated by cold and dry conditions, while during a positive NAO 439 mode, mild and humid conditions prevail (Fig. 11A; Hurrell and van Loon, 1997; Marshall et 440 al., 2001). This NAO-temperature link is reflected in  $\delta^{18}$ O of precipitation (Baldini et al., 441 2008; Mischel et al., 2015; Comas-Bru et al., 2016) and thus can be reconstructed using 442 suitably sensitive speleothems. 443

444 To quantify the effect of the NAO on regional weather and climate, precipitation and 445 temperature records of the GNIP stations Bad Salzuflen, Emmerich, Koblenz, Wasserkuppe

Rhön, and the meteorological stations Hagen-Fley and Hemer were averaged for the months 446 December to March. December-March average NAO index values (NAO<sub>DIFM</sub>) were 447 calculated using data provided by the Koninklijk Nederlands Meteorologisch Instituut 448 (KNMI; http://www.knmi.nl/). Both precipitation amount ( $r \ge 0.36$ ,  $p \le 0.05$ ,  $n \ge 19$ , except 449 Hemer) and temperature ( $r \ge 0.58$ ,  $p \le 0.03$ ,  $n \ge 5$ ; except Wasserkuppe/Rhön) correlate 450 significantly with the NAO<sub>DIFM</sub> for most stations (Fig. 11). Thus, precipitation amount and 451 temperature above Bunker Cave are affected by the NAO during winter months. Since both 452 parameters are imprinted on drip rate and the dripwater isotope composition, it is possible to 453 reconstruct NAO variability using proxy time-series from speleothems of Bunker Cave. As 454 the precipitation in this region is equally distributed throughout the year, substantial summer 455 precipitation between April to November - despite evapotranspiration - may weaken the 456 imprint of the winter NAO signal on speleothems (Mischel et al., 2015). However, studies by 457 458 Baldini et al. (2008) and Comas-Bru et al. (2016) show that many Central European regions are sensitive to reconstruct NAO variability. A moderate connection between  $\delta^{18}$ O and NAO 459 variability has also been detected for Herbstlabyrinth-Adventshöhle in Central Germany 460 (Mischel et al., 2015), supporting the notion that speleothems from these caves record NAO 461 variability. 462

With short residence times of <3 years and limited signal smoothing in the overlying epikarst, Bunker Cave proves to be a sensitive site for high-resolution proxy reconstructions of past climate dynamics under the influence of North Atlantic atmospheric circulation. The links between Bunker Cave hydrology and the NAO found in this study are consistent with the interpretation brought forward by previous workers (Fohlmeister et al., 2012; Wassenburg et al., 2016).

469

470 5.4 Wider implications of the present data set for speleothem research in general

In case of Bunker Cave, Mg/Ca ratios as well as  $\delta^{13}$ C values depend on drip rate and thus, on precipitation and infiltration amount (Riechelmann et al., 2011, 2013; Fohlmeister et al., 2012). Therefore, both proxies are well-suited for reconstructions of precipitation and NAO variability.

Disentangling air temperature based on speleothem  $\delta^{18}$ O is a more complex task. 476 There is a clear temperature signal in dripwater  $\delta^{18}$ O of all drip sites. This temperature signal 477 of speleothem  $\delta^{18}$ O, however, is altered by (drip rate-related) fractionation processes during 478 calcite precipitation in the cave (Riechelmann et al., 2013). In the case of speleothem  $\delta^{18}$ O in 479 Bunker Cave, variations in both parameters (precipitation and temperature) are recorded 480 481 (Fohlmeister et al., 2012). A proxy not recorded in speleothem calcite but in dripwater is δD showing a well-defined temperature signal. Fluid inclusions extracted from speleothems could 482 be used to quantify  $\delta D$  and  $\delta^{18}O$  of the original dripwater and to constrain past temperature 483 changes (McGarry et al., 2004; van Breukelen et al., 2008; Affolter et al., 2015). Given that 484 the temperature pattern is unaltered in dripwater  $\delta^{18}$ O, fluid inclusions  $\delta^{18}$ O and  $\delta$ D analyses 485 486 should allow for the reconstruction of past temperature variability from speleothems in Bunker Cave and thus, variations of NAO strength. 487

The most striking observation of the present study is the difference between hydraulic response time, i.e. immediate drip rate response (<1 year; Fig. 7), and the transfer time of water from the soil to the cave (ca. 2.5 years; Fig. 10). While drip rate response to annual precipitation occurs without significant lag on (sub-)annual basis, the temperature signal is delayed by up to 2.5 years. Thus, the precipitation and temperature signals recorded by different proxies in speleothems are not fully synchronous. In case of Bunker Cave this delay is negligible, because here speleothems record proxies on decadal rather than annual or 495 seasonal scales (Riechelmann et al., 2011; Fohlmeister et al., 2012). Such proxy biases to
496 either water transfer or hydraulic response time must be considered in (sub-)annual
497 reconstructions, and where the signal transfer times for both parameters differ.

498

499 **6.** Conclusions

500

The seven year-long monitoring of hydrological parameters in and above Bunker Cave, NW Germany, provides unprecedented insights into processes influencing  $\delta^{18}$ O and  $\delta$ D in dripwater and speleothems that cannot be resolved with shorter-term monitoring programmes. The results of this study aim at quantifying the most relevant processes that may alter isotope proxies in Bunker Cave speleothems over time scales of several years:

Inter-annual drip rate dynamics are controlled by annual infiltration and thus, annual
 precipitation. No lag of the hydraulic response time is observed at annual resolution.

508 2. Water transfer time is 3 months for a 40 cm-thick soil layer and 4 months for a 70 cm-thick
509 soil. It takes approximately 2.5 years for meteoric water to reach the cave chambers.

510 3. Neither oxygen nor hydrogen isotopes of dripwaters are altered and hence, record the 511 temperature signal of meteoric precipitation. As a consequence,  $\delta^{18}$ O and  $\delta$ D from fluid 512 inclusions in speleothems are genuine temperature proxies at this site.

4. Temperature and precipitation during winter months (December-March) are significantly
influenced by the NAO. Hence, long-term NAO variations are recorded in speleothems
from northwestern Germany.

5. Monitoring data reveal a difference between the hydraulic response time and the water
transfer time. This, however, does not affect the proxy interpretation of speleothems from
this cave, because of their slow growth rate. But, with regard to other caves, especially

with seasonally to annually resolved speleothems, this time offset may lead to difficultiesregarding the interpretation of various proxy data.

521 6. This study provides insights into multi-annual processes operating in cave depositional
522 environments that are of relevance in speleothem research and can serve as a template for
523 other cave monitoring programmes.

524

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526

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537

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- 783

#### 784 Figure Captions

785

Fig. 1 Geological map of the northern Rhenish Slate Mountains in northwestern Germany.
Locations of Bunker Cave, German Cave Museum Iserlohn and the meteorological stations
(MS) Hagen-Fley and Hemer are shown (modified after Riechelmann et al., 2011).

Fig. 2 Plan view of Bunker Cave with the locations of drip sites TS 1 and TS 5 in chamber 1
and TS 2, TS 3 and TS 8 in chamber 2 (modified after Scheudoschi and Hasenkamp in
Scheudoschi, 2007).

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Fig. 3 Monthly values of temperature as well as rainwater oxygen and hydrogen isotope data 794 and mean annual values of precipitation, infiltration, temperature as well as rainwater oxygen 795 and hydrogen isotope data. A) Precipitation data from 2000 to 2006 of the meteorological 796 797 station (MS) Hagen-Fley and data from 2007 to 2013 measured during this study at the German Cave Museum Iserlohn. Temperature data from 2000 to 2007 were provided by the 798 MS Hagen-Fley and 2008 to 2013 by the MS Hemer. Infiltration data are calculated using 799 data of both meteorological stations. B) Rainwater oxygen and hydrogen isotope data 800 measured from the rain sampled at the German Cave Museum Iserlohn. 801

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Fig. 4 A) Comparison of mean annual oxygen isotope data of four GNIP stations with rainwater data collected during this study. B) Comparison of mean annual hydrogen isotope data of four GNIP stations with rainwater data collected in this study. C) Local meteoric water line (LMWL) of the rainwater and position of the dripwaters on the LMWL (small box).

807

Fig. 5 Mean annual drip rates in Bunker Cave. Both manual and automatic drip rate data areshown.

810

Fig. 6 Monthly and mean annual oxygen and hydrogen isotope data of soil- and dripwaters.

813	Fig. 7 A) Correlation coefficients between mean annual drip rate and annual infiltration. B)
814	Correlation coefficients of mean annual drip rate with annual precipitation. Note the offset to
815	obtain the hydraulic response time of the reservoir to precipitation and infiltration.
816	
817	Fig. 8 Drip characteristics of drip site TS 8 according to Smart and Friederich (1987) and
818	Baker et al. (1997). Data from 2013 are not displayed as they do not cover the whole year.
819	
820	Fig. 9 Correlation coefficients of monthly $\delta^{18}$ O and $\delta$ D values with atmospheric temperature
821	for both soilwater sites BW 1 and BW 2.
822	
823	Fig. 10 Correlation coefficients between the monthly $\delta^{18}O$ and $\delta D$ values of all dripwaters and
824	atmospheric temperature.
825	
826	Fig. 11 A) NAO index for December to March calculated for the period 1975-2013

(http://www.knmi.nl/). B) Precipitation data of the GNIP and meteorological stations for
December to March for the period 1975-2013. C) Temperature data of the GNIP and
meteorological stations for December to March for the period 1978-2013.

1	Sensitivity of Bunker Cave to climatic forcings highlighted through multi-annual
2	monitoring of rain-, soil-, and dripwaters
3	
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22	Keywords: drip rate, infiltration, temperature, oxygen and hydrogen isotopes, NAO, cave
23	microclimate monitoring
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### 26 ABSTRACT

The last two decades have seen a considerable increase in studies using speleothems as 27 archives of past climate variability. Caves under study are now monitored for a wide range of 28 environmental parameters and results placed in context with speleothem data. The present 29 study investigates trends from a seven year long monitoring of Bunker Cave, western 30 Germany, in order to assess the hydraulic response and transfer time of meteoric water from 31 32 the surface to the cave. Rain-, soil-, and dripwater were collected from August 2006 to August 2013 at a monthly to bimonthly resolution and their oxygen and hydrogen isotope 33 composition was measured. Furthermore, drip rates were quantified. Due to different drip 34 characteristics, annual mean values were calculated for the drip rates of each drip site. 35 Correlations of the annual mean drip rate of each site with precipitation and infiltration 36 demonstrate that the annual infiltration, and thus the annual precipitation control the inter-37 38 annual drip-rate variability for all except one site. The hydraulic response is not delayed. All drip sites display identical long-term trends, which suggests a draining of a common karst 39 reservoir over these seven years of monitoring. Correlations of soil- and dripwater monthly 40  $\delta^{18}$ O and  $\delta$ D values with atmospheric temperature data reveal water transfer times of 3 41 months for soilwater at site BW 2 (40 cm depth) and 4 months for 70 cm depth (soilwater at 42 site BW 1). Finally, the water reaches the cave chambers (15 to 30 m below land surface) 43 after ca. 2.5 years. Consequently, a temporal offset of 29 to 31 months (ca. 2.5 years) between 44 the hydraulic response time (no time lag on annual basis) and the water transfer time (time lag 45 of 29 to 31 months) was found, which is negligible with regard to Bunker Cave speleothems 46 because of their slow growth rates. Variations in drip rate and thus, precipitation and 47 infiltration are recorded by  $\delta^{13}$ C and Mg/Ca ratios in speleothem calcite. Speleothem  $\delta^{18}$ O 48 values reflect both temperature and precipitation signals due to drip rate-related fractionation 49 processes. We document that long-term patterns in temperature and precipitation are recorded 50

in dripwater patterns of Bunker Cave and that these are linked to the North AtlanticOscillation (NAO).

53

#### 54 **1. Introduction**

55

Palaeoclimate reconstructions based on speleothems, i.e. mostly carbonate deposits 56 formed in caves, have increased significantly during the last two decades (for a summary, see 57 Fairchild and Baker, 2012). The most important strengths of speleothems are the precise 58 <sup>230</sup>Th/U dating (e.g., Dorale et al., 2004; Scholz and Hoffmann, 2008; Cheng et al., 2013) and 59 the availability of several, mostly geochemical, parameters such as carbon and oxygen isotope 60 values, major and trace elemental abundances, and  $\delta^{18}O$  and  $\delta D$  of fluid inclusions (e.g., 61 Niggemann et al., 2003; Mangini et al., 2005; Vonhof et al., 2006; Fohlmeister et al., 2012; 62 Scholz et al., 2012; Luetscher et al., 2015). These can be used for single- or multi-proxy 63 approaches to reconstruct past climate dynamics. 64

In order to gain a better understanding of the processes influencing geochemical 65 proxies in the soil and epikarst zone, as well as processes acting during deposition of 66 speleothems, sophisticated monitoring programmes have been established (e.g., Spötl et al., 67 2005; Mattey et al., 2008b, 2016; Riechelmann et al., 2011, Wassenburg et al., 2013; Genty et 68 al., 2014; van Rampelbergh et al., 2014, Breitenbach et al., 2015; Treble et al., 2016). In the 69 context of these efforts, cave air temperature, pCO<sub>2</sub> and humidity, drip rate, as well as the 70 isotopic composition of rain-, soil- and dripwater and the element concentrations of soil-, and 71 dripwater have been recorded. Furthermore, in order to link the dripwater with speleothems 72 used for palaeoclimate reconstructions, recent cave carbonate precipitates have been 73 investigated in combination with their respective dripwater composition and the hydraulic 74

regime in the cave (e.g., Miorandi et al., 2010; Tremaine et al., 2011; Riechelmann et al.,
2013, 2014).

Most monitoring studies focused on seasonal variations of the above-mentioned 77 parameters, whereby, for example, the drip rate was analysed to study the response to rainfall 78 events or the hydrological connection of the drip sites (Baker et al., 1997; Mattey et al., 79 2008b; Riechelmann et al., 2011). The analysis of dripwater  $\delta^{18}$ O and  $\delta$ D values shows either 80 81 seasonal variations (Mattey et al., 2008a, Breitenbach et al., 2015) or rather stable values close to the annual mean of rainwater (Riechelmann et al., 2011; van Rampelbergh et al., 82 83 2014). Seasonal variations in cave dripwater reflect a fast transfer of the water (e.g., Breitenbach et al., 2015). The lack of an intra-annual pattern points to strong mixing in the 84 epikarst and/or the vadose zone with transfer times between the soil and the cave drip site in 85 excess of one year (e.g., Riechelmann et al., 2011; van Rampelbergh et al., 2014). Financial 86 constraints, accessibility, or lack in manpower commonly limit monitoring studies to 87 durations of a few years or less. Thus far, only a very limited number of studies focused on 88 multi-annual trends of the monitored parameters and implications for speleothem research 89 (Genty and Deflandre, 1998; Treble et al., 2013; Genty et al., 2014, Breitenbach et al., 2015; 90 Mattey et al., 2016). To the knowledge of the authors, only the study by Genty et al. (2014) 91 92 detrended the seasonal signal in order to gain insights in potential longer-term (> 5yrs) trends. This is of significance as particularly the longer-term trends are relevant for the assessment of 93 proxy data from speleothems recording decadal or longer variability only. 94

This paper documents and discusses observations of precipitation/infiltration and drip rate as well as atmospheric temperature and the oxygen and hydrogen isotopic composition of rain-, soil-, and cave dripwater from a seven year-long monitoring campaign in Bunker Cave in northwestern Germany. The main goals of the present study are: (i) to quantify the longerterm (multi-annual) variability of the environmental parameters; (ii) to assess the response time of the carbonate precipitating drip sites to these parameters; (iii) to identify signal
smoothing and possible alteration processes during percolation through the epikarst; and (iv)
to draw, where possible, general implications for speleothem research.

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## 104 2. Climate and cave parameters

105

The climate of northwestern Germany is warm-temperate, i.e. fully humid (equally distributed rainfall amount throughout the year) with warm summers (Kottek et al., 2006). Bunker Cave is located between the villages of Iserlohn and Letmathe in the Rhenish Slate Mountains in the NW part of the Sauerland, Germany (Fig. 1). It is part of the Bunker-Emst-Cave system (3.5 km long) with the Bunker Cave entrance (51°22'N, 07°40'E) being located at 184 m above sea level (asl) on a south-facing hill slope of the Dröscheder Emst karst plateau (Grebe, 1993; Hammerschmidt et al., 1995).

The host rock consists of Middle to Upper Devonian massive limestone (von Kamp, 113 1972). The rock overburden of the cave is between 15 and 30 m (Grebe, 1993) and the host 114 rock is overlain by ca. 70 cm inceptisol to alfisol (USDA Soil Taxonomy). The colour varies 115 between dark and yellowish brown (10YR 3/3 and 10YR 5/6) for the upper soil layers and 116 bright reddish brown to bright brown (5YR 5/8 to 7.5YR 5/8) for the lower layer (Munsell 117 soil colour charts). The vegetation above the cave consists of deciduous forest (mainly ash 118 and beech trees) and scrubs (Riechelmann et al., 2011). Bedding dips to the North or 119 Northwest (von Kamp and Ribbert, 2005) and water percolates mainly along fractures and 120 bedding planes. This feature and the fact that Bunker Cave is located in a south-facing hill 121 reduces the effective catchment of the cave dripwater to a few hundred  $m^2$ . Furthermore, the 122 Dröscheder Emst karst plateau is partly used as a residential area and ca. 15 to 20% of the 123 catchment area is anthropogenically sealed. Furthermore, a railway route runs above the cave. 124

# 126 **3. Materials and Methods**

127

### 128 3.1 Monitoring and sample collection of rain-, soil- and dripwater

129

The monitoring programme performed in and above Bunker Cave ran from August 2006 to August 2013. Rainwater samples were collected with a rain gauge according to the DIN 58666C norm on the roof of the German Cave Museum Iserlohn (51°22'N, 07°38'E; 175 m asl), located 1.5 km from Bunker Cave (Fig. 1). Rainfall amount was measured and sampled daily and collected as monthly samples between 2007 and 2012. In 2013, rainwater samples were collected only bimonthly. Rainwater samples were stored in PET bottles in a fridge to minimize evaporation.

Two soilwater sampling sites were installed above Bunker Cave. The soilwater suction probes were manufactured by Umwelt-Geräte-Technik GmbH (UGT, Germany). One probe (BW 1) was installed at a depth of 70 cm and water was sampled monthly between 2007 and 2011. The second probe (BW 2) sampled water at 40 cm depth in monthly intervals between 2009 and 2011. In 2012 and 2013, both soilwater sites were sampled only bimonthly.

In total, five drip sites (TS 1, TS 2, TS 3, TS 5 and TS 8; Fig. 2) were monitored in 142 Bunker Cave from 2006 to 2013. Sampling at drip site TS 8 started in 2007. Drip sites TS 1 143 and TS 5 are located in chamber 1. All other sites are located in chamber 2 of Bunker Cave 144 (Fig. 2). Dripwater samples were integrated over one month in order to obtain sufficient 145 volumes of water for multi-proxy geochemical analyses. These monthly samples were taken 146 between 2006 and 2011, while bimonthly samples were taken between 2012 and 2013. An 147 exception is drip site TS 1, where sufficient water could be collected during each visit. 148 Dripwater first dripped onto a drip counter placed in a plastic box and was then transferred 149

150 into a closed bottle via tubing to minimize  $CO_2$  degassing and potential evaporation. 151 Comparison of instantaneous and monthly collected water revealed similar  $\delta^{18}O$  values, 152 confirming that fractionation processes due to evaporation are negligible (Riechelmann et al., 153 2011). For further details of the monitoring, the reader is referred to Riechelmann (2010) and 154 Riechelmann et al. (2011; 2012a; 2013; 2014).

Drip rates were measured manually with a stopwatch. The following equation was used to convert drip rate into volumetric discharge (*z*):

157

158  $z \,[\text{ml/time}] = \text{drip rate [drips/time]}^* V \,[\text{ml/drips}]$  (1)

159

where V is the volume of each drip. The volume of a drip was determined by a defined 160 collection time, the total amount of water and the drip rate (in drips per min). In some cases, 161 the drip rate at site TS 8 was not quantified by means of a stopwatch, and was instead 162 calculated via the amount of an instantaneous water sample (collected during the monitoring 163 tour) and the known drip volume (determined as described above). Furthermore, drip rates 164 were measured automatically by acoustic drip counters (Stalagmate; Mattey and Collister, 165 2008), which were placed under drip sites TS 2 (2009-2013), TS 3 (2010-2013), TS 5 (2007-166 2013) and TS 8 (2009-2013). 167

168

# 169 3.2 $\delta^{l8}O$ and $\delta D$ of rain-, soil- and dripwater

170

The oxygen and hydrogen isotopic compositions of all water samples were determined at the University of Innsbruck. Water samples were collected in small, airtight glass vials without headspace. The oxygen isotope composition was analysed using a ThermoFinnigan DELTA<sup>plus</sup>XL mass spectrometer connected with a Gasbench II using the CO<sub>2</sub> equilibrium technique. The 1σ reproducibility of the  $\delta^{18}$ O values is ±0.09 ‰ VSMOW (Spötl et al., 2005). The hydrogen isotope composition was measured using a ThermoFinnigan DELTA<sup>plus</sup>Advantage equipped with a TC/EA high-temperature pyrolysis unit. The 1σ reproducibility is ±1 ‰ VSMOW.

179

# 180 *3.3 Evapotranspiration and infiltration*

181

Instrumental climate data of two nearby meteorological stations (Fig. 1) - Hagen-Fley
(51°25'N, 07°29'E; 100 m asl; 2000-2007; www.dwd.de) and Hemer (51°23'N, 07°45'E; 200
m asl; 2008-2013; www.meteomedia.de) - were used to calculate the evapotranspiration after
Haude (1955) using the following equation:

186

187 
$$E_{pot} [mm/day] = x [mm/day * hPa] * P^{13} [hPa] * (1 - (F^{13} [\%] / 100))$$
 (2)

188

189 where  $E_{pot}$  is the potential evaporation, *x* represents the monthly coefficient depending on 190 vegetation,  $P^{13}$  is the saturation pressure at 1 p.m. and  $F^{13}$  is the relative humidity at 1 p.m.. 191 The saturation pressure was calculated using the Magnus-formula:

192

193 
$$P^{I3}$$
 [hPa] = 6.107 [hPa] \*10^((7.5 \*  $T^{I3}$  [°C]) / (237 +  $T^{I3}$  [°C])) (3)

194

195  $T^{I3}$  is the air temperature at 1 p.m.. Equations 2 and 3 were used to calculate the amount of 196 water which can potentially infiltrate ( $Inf_{pot}$ ) into the soil and thus, into the cave. Therefore, 197 the following equation was used:

199 
$$Inf_{pot} [mm/day] = N [mm/day] - E_{pot} [mm/day]$$
 (4)

where *N* is precipitation. The amount of water infiltrating into the soil above Bunker Cavewas determined using precipitation data from the German Cave Museum Iserlohn.

203

204 *3.4 GNIP stations* 

205

Data of four nearby GNIP (Global Network of Isotopes in Precipitation; http://wwwnaweb.iaea.org/napc/ih/IHS\_resources\_gnip.html) stations were used for comparison with monitoring and meteorological data. These include Bad Salzuflen (52°06'N, 08°43'E; 100 m asl), Emmerich (51°49'N, 06°36'E; 43 m asl), Koblenz (50°21'N, 07°34'E; 97 m asl) and Wasserkuppe Rhön (50°30'N, 09° 57'E; 921 m asl).

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212 4. Results
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213

Seasonally resolved monitoring data of rain-, soil- and dripwater of Bunker Cave were 214 215 partly published by Immenhauser et al. (2010), Kluge et al. (2010, 2013), Riechelmann et al. (2011, 2012a, b, 2013, 2014), Fohlmeister et al. (2012), Münsterer et al. (2012), and 216 Wackerbarth et al. (2012). In this study, we present monthly data as well as annual mean 217 values of the relevant parameters, which were calculated using the monthly or bimonthly 218 values of the respective year to (i) examine long-term variability and (ii) compare these data 219 with precipitation/infiltration and temperature data to determine the hydraulic response time 220 and water transfer time. Annual means were obtained from monthly data (January to 221 December). 222

```
224 4.1 Precipitation, infiltration, temperature and rainwater \delta^{18}O and \delta D
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226 For this study, the time period from 2000 to 2013 is considered. Annual mean values are shown for all years except 2013, since monitoring stopped in August of this year. Hence, 227 all mean values for 2013 are related to this time interval. Furthermore, annual mean values for 228 2006 were not calculated, due to insufficient data. The lowest annual precipitation sum (720 229 mm) was recorded in 2003, without considering 2013 with the lowest precipitation sum of 230 231 447 mm. The highest annual rainfall (1202 mm) and the highest mean annual infiltration (546 mm) occurred in 2007 (Fig. 3A). The lowest annual mean infiltration was observed in 2003 (-232 171 mm). Infiltration at Bunker Cave is lower during summer months than during winter 233 234 (Riechelmann et al., 2011). Monthly temperature data show low values in winter and high ones during summer. The lowest mean annual air temperature (7.1°C) was observed in 2010, 235 while the highest (11.3°C) was observed twice, in 2000 and 2007 (Fig. 3A). 236

Oxygen and hydrogen isotope values of monthly rainwater samples display a seasonal variability with higher values during summer and lower ones during winter months (Fig. 3B). In the years 2010 and 2013, the lowest mean annual  $\delta^{18}$ O and  $\delta$ D values of rainwater were observed ( $\delta^{18}$ O: -9.20 and -9.01‰;  $\delta$ D: -66.2 and -62.5‰), while the highest mean annual values were observed in 2011 ( $\delta^{18}$ O: -5.92‰;  $\delta$ D: -45.8‰; Fig. 3B). Monthly  $\delta^{18}$ O and  $\delta$ D values of rainwater are highly correlated (r = 0.98, p < 0.001; n = 65; see also Fig. 4C).

Since monthly rainwater samples collected during the monitoring do not cover the same time period as the monthly samples from the GNIP stations, mean annual values are used here for comparison. Mean annual oxygen and hydrogen isotope values of rainwater collected at the German Cave Museum correlate well with those of nearby GNIP stations (Figs. 4A, B). The Local Meteoric Water Line (LMWL) of the rainwater (Fig. 4C) is close to that of the GNIP station Bad Salzuflen ( $\delta D = 7.72 * \delta^{18}O + 5.7$ ; r = 0.98; Stumpp et al., 2014). Thus, rainwater collected during the monitoring period reflects the climate conditions of western Germany. The slope of 7.72 is close to that of the Global Meteoric Water Line (8), indicating insignificant evaporation of the collected samples (Gat et al., 2000). Since all dripwater data plot on the LMWL (Fig. 4C) evaporation in the soil, epikarst and cave generates no significant imprint on their corresponding isotope values. Furthermore, both isotope systems depend on the regional surface air temperature and the isotopic composition of Bunker Cave dripwater reflects the infiltration weighted mean annual  $\delta^{18}$ O and  $\delta$ D values of rainwater (Riechelmann et al., 2011).

257

258 *4.2 Drip rate* 

259

Due to different drip characteristics of the different drip sites (ranging from seepage 260 flow to seasonal drip; Riechelmann et al., 2011; Appendix S1 and Fig. 8), annual mean drip 261 rates were calculated and their long-term variability compared. Both manually and 262 automatically obtained mean annual drip rates show the same trends (Fig. 5). Sites TS 1, TS 2 263 and TS 5 show a decreasing trend in mean drip rates over the monitoring period, which is also 264 reflected in cross-correlations of mean annual drip rates at these sites (TS 1 vs. TS 2: r = 0.82; 265 TS 1 vs. TS 5: r = 0.85; TS 2 vs. TS 5: r = 0.88;  $p_{all} \le 0.02$ ,  $n_{all} = 7$ ). The annual mean drip 266 rate at site TS 3 correlates only with the annual mean drip rate at site TS 2 within the 95% 267 confidence level (r = 0.81) and with TS 1 (r = 0.79) and TS 5 (r = 0.72) just below the 95% 268 confidence level  $(n_{all} = 6)$ . However, it can be assumed that the annual mean drip rate at site 269 TS 3 follows the same trend as sites TS 1, 2 and 5. At drip site TS 8 a similar annual mean 270 drip rate was observed in 2008 and 2009 before drip rates increased until 2011, followed by a 271 decreasing trend. Drip rate of drip site TS 8 does not show any significant correlation with 272 any other drip rate. 273

Seasonal trends are difficult to identify for soilwater, because of lack of data (no 277 water), especially summer months. An exception is soilwater at site BW 1 which yielded a 278 complete time series from 2007 to 2008. There, higher  $\delta^{18}$ O and  $\delta$ D values occur during 279 summer/autumn months, while lower values are observed in winter/spring months (Fig. 6). 280 The lowest mean annual  $\delta^{18}$ O and  $\delta$ D values occurred in 2010 and 2011 for both soilwaters, 281 while the highest values are shown in 2007 and 2008 (Fig. 6). Monthly  $\delta^{18}O_{BW1}$  values are 282 significantly correlated with  $\delta^{18}O_{BW2}$  values (Table 1). The same can be observed for  $\delta D$ 283 (Table 2). 284

Seasonal trends in  $\delta^{18}$ O and  $\delta$ D of dripwaters are minor, but most pronounced for 285 water at site TS 1. In general, lower isotope values occur in summer, while higher values are 286 observed in winter (Fig. 6). Correlations between  $\delta^{18}$ O and  $\delta$ D values of monthly samples of 287 the same site are significant for all dripwaters and both soilwater samples (r > 0.44; p < 0.001; 288  $n \ge 21$ ). All dripwaters share similarities in their oxygen and hydrogen isotopic composition 289 as shown by positive cross-correlations (Tables 1 and 2). Mean annual values display the 290 overall trend for  $\delta^{18}$ O and  $\delta$ D, which is similar for all drip sites considering the errors (Fig. 6). 291 Oxygen and hydrogen isotope values increased until 2009, reached a plateau and decreased 292 around 2012. This trend is less distinct for dripwater at site TS 8 (Fig. 6). 293

294

# 295 5. Interpretation and Discussion

296

The positive correlations of mean annual drip rates of all sites except TS 8 (Fig. 5) 299 suggests that: (i) these sites are characterized by similar hydrological properties (Baker et al., 300 1997), and (ii) the drip rates of these sites most likely respond to the same environmental 301 302 (climate) forcing. According to Riechelmann et al. (2011), no drip site showed a direct response to rainfall events, but rather a lagged response of up to several months due to slowly 303 304 increasing hydrological pressure in the karst reservoir above the cave. Correlations between 305 drip rates and precipitation and infiltration during the preceding years based on monitoring and meteorological data of the stations Hagen-Fley and Hemer were used to determine the 306 response time of mean annual drip rates to mean annual precipitation and infiltration. The 307 308 correlation coefficients (r) between drip rate and infiltration show that the mean annual drip rates at TS 1 and TS 3 are controlled by mean annual infiltration of the same year, as observed 309 for other cave systems (Fig. 7A; Genty and Deflandre, 1998; Baker et al., 2000; Miorandi et 310 311 al., 2010; Tremaine and Froelich, 2013). Correlation coefficients at drip sites TS 2 and 5 are close to the 95% confidence level. Since the infiltration amount is controlled by precipitation 312 313 amount and temperature (water loss due to evapotranspiration), correlations between mean 314 annual drip rate and annual precipitation amount were calculated (Fig. 7B). Positive correlations at the 95% confidence level were found for TS 2 and TS 3 for the same year 315 interval, while correlation coefficients of TS 1 and 5 are just below the 95% confidence level. 316 Several sources of error must be considered, however, which lead to a lack of correlation at 317 the 95% confidence level. Lack of data (< 12 for annual means) may be the problem in case of 318 drip site TS 5, since neither infiltration nor precipitation correlate at the 95% confidence level 319 with drip rate. This might also be the case for sites TS 1 and 2. The calculated mean annual 320 drip rate of the former might be biased, since it shows pronounced seasonal drip rate 321 variability, with most changes not being covered by the monitoring (Figs. 5 and S1 in 322 supplementary material). Finally, the calculated mean annual infiltration in the soil reflects 323

the *potential* rather than *effective* infiltration. Furthermore, it must be kept in mind that effective infiltration is biased towards winter (Wackerbarth et al., 2010; Riechelmann et al., 2011), which might affect the correlation.

A decrease of the drip rate was already observed for two other drip sites and TS 2 in Bunker Cave for the first three years as reported in Riechelmann et al. (2011). As shown here, this trend continued (Fig. 5), i.e. the reservoir was drained further during the seven years. This might be due to relatively dry years with low infiltration at the beginning of the century. The following years (2007 to 2010) with higher infiltration most likely did not fill the reservoir sufficiently to stop the draining and afterwards infiltration decreased again (2011 to 2013; Fig. 3A).

The mean annual drip rate at site TS 8 shows a significant positive correlation with mean annual infiltration with a lag of three years, but no significant correlation near the 95% confidence level with precipitation. Drip site TS 8 is the only site, whose drip rate does not show the same trends as the other sites and also lacks a correlation with infiltration and precipitation for the interval recorded by the other drip sites. This absence of any correlation might be due to a change in drip behaviour during the monitoring period.

The drip characteristics of all drip sites, plotted according to Smart and Friederich 340 (1987) and Baker et al. (1997; see Figs. 8 and S1 in supplementary material to this paper) 341 highlight minor variability, with more or less the same drip characteristics over time. Drip 342 behaviour changed drastically at TS 8 during the monitoring period (Fig. 8): while seasonal 343 drip characteristics prevailed in 2008 and 2009, this drip shows seepage flow behaviour 344 between 2010 and 2012. Possible reasons for this change in drip characteristics include: (i) the 345 flow paths and the reservoir behaviour in the epikarst underwent changes as a result of 346 strongly variable rainfall events (Tooth and Fairchild, 2003), (ii) the flow paths of percolating 347 water changed or became blocked due to prior calcite precipitation (PCP; Fairchild et al., 348

2006) occluding the flow paths of water; (iii) the overhanging stalactite was broken off 349 (earthquake, human interference etc.) resulting in drip parameter changes (Baldini et al., 350 2006). Amongst these scenarios, it is conceivable that rainfall filled the reservoir feeding TS 8 351 to a threshold resulting in a more continuous water flow into the cave. PCP is a more likely 352 explanation for the change in drip behaviour as documented for this site (Riechelmann et al., 353 2011). The stalactite was removed in June 2008, but the change in drip rate lagged by one 354 year, suggesting that the drip pattern was not controlled by the stalactite and thus the third 355 option mentioned above is considered unlikely. Concluding, the drip rate at site TS 8 is 356 controlled by infiltration and hence rainfall, but the dripwater signal reflecting both of these 357 parameters is strongly affected by prior calcite precipitation in the aquifer. Although the 358 catchment of the cave is partly sealed, infiltration and thus the precipitation signal can still be 359 recognized in the drip rate. 360

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### 362 5.2 Constraints on the water transfer time

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Water transfer times from the surface to the cave are highly variable and range from 364 days to several years not only for different caves but also for different drip sites in a given 365 cave (e.g., Kaufmann et al., 2003; Mattey et al., 2008b, Lambert and Aharon, 2010; Treble et 366 al., 2013; Genty et al., 2014, Breitenbach et al., 2015). Here, the dependency of rainwater 367  $\delta^{18}$ O and  $\delta$ D on temperature ( $\delta^{18}$ O versus T: r = 0.73;  $\delta$ D versus T: r = 0.68;  $n_{both} = 67$ ;  $p_{both} < 100$ 368 0.001; Riechelmann et al., 2011) is used to quantify the water transfer time. Correlation 369 coefficients (r) between  $\delta^{18}$ O and  $\delta$ D of soil- and dripwater with surface air temperature were 370 calculated using data of the meteorological stations Hagen-Fley and Hemer. To this end, 371 monitoring data were correlated with temperature data of preceding years. Since rainwater 372

373  $\delta^{18}$ O and  $\delta$ D values correlate positively with temperature, only positive correlations are 374 relevant to determine the water transfer time.

Monthly  $\delta^{18}$ O and  $\delta$ D values of soilwater site BW 1 at 70 cm correlate significantly 375 with the atmospheric temperature with a lag of four months (Fig. 9). In case of soilwater site 376 BW 2 a significant correlation was found using a three months lag (40 cm; Fig. 9). However, 377 gaps in the monthly soilwater data during dry summer months limit the significance of these 378 calculations and they should therefore be treated with caution. Thus, this is also resulting in 379 other highly significant correlations at 63 months lag time in case of BW 2. Since  $\delta^{18}O$  and 380  $\delta D$  values of soilwater correlate significantly with atmospheric temperature at both sites, and 381 monthly  $\delta^{18}$ O and  $\delta$ D values also correlate for both soilwater sites, we conclude that water 382 isotopes are not or only slightly altered by processes in the soil zone such as evaporation and 383 uptake of biogenic CO<sub>2</sub> into the water (e.g., Lachniet, 2009). Thus, the temperature 384 dependency of rainwater is preserved in the  $\delta^{18}$ O and  $\delta$ D values of soilwater above Bunker 385 Cave. 386

Because dripwater at site TS 1 was collected as an instantaneous sample, the  $\delta^{18}$ O and 387  $\delta D$  values are not representative for the whole month as is the case for the other dripwaters. 388 Therefore, calculations with monthly data were only performed for all other drip sites 389 resulting in a lag of 29 to 31 months (Fig. 10). Difficulties occurred for TS 2, TS 3, and TS 8 390 in case of correlations using  $\delta^{18}$ O. There, the highest correlations are found between 41 and 391 66 months and only slightly lower ones are observed for a lag of 29 to 31 months. 392 Calculations using  $\delta D$  of all drip sites except TS 2 and  $\delta^{18}O$  for TS 5 do not show these 393 features, which are most likely due to different mixing of the younger and older water in the 394 reservoir as a consequence of different transfer times. Variability in the transfer times 395 between different drip sites may arise due to: (i) variable thickness of the soil layer above 396

Bunker Cave, and (ii) differential thickness of the host rock overlying the cave, as well as (iii) 397 differential lengths and types of individual flow paths (e.g., Tooth and Fairchild, 2003). The 398 water retention capacity of soil is another factor that may or may not lead to an admixture 399 waters with different residence time in the soil and epikarst zone (Hölting and Coldewey, 400 2013). Following dry periods, the portion of older water in the soil increases (Kottek et al., 401 2006) and the opposite is found during more humid periods. Although, rainfall is equally 402 distributed throughout the year, infiltration is not, as it is lower during summer than during 403 winter months (Wackerbarth et al., 2010; Riechelmann et al., 2011). Thus, water retention 404 capacity might have a small effect on water transfer times at Bunker Cave sulphate and nitrate 405 data show that dripwaters at sites TS 1 and TS 5 have shorter residence times compared to 406 sites TS 2, 3, and 8 (Riechelmann et al., 2011). This might explain why both  $\delta^{18}O$  and  $\delta D$ 407 correlations with temperature performed well for TS 5. Drip water at TS 2 appears to be the 408 site with the longest and strongest buffering in the epikarst zone. In contrast to most others 409 this site displays seepage flow characteristics (see Appendix S1) and the oldest age based on 410 tritium data (Riechelmann et al., 2012a). This pattern might explain why the correlations at 411 this site are weakest. However, the shift in correlations using  $\delta D$  compared to the other drip 412 sites (Fig. 10) remains insufficiently explained. Since  $\delta^{18}$ O and  $\delta$ D values of dripwater at site 413 TS 1 correlate with those of all other drip sites it can be assumed that the TS 1 water has a 414 similar transfer time of 29 to 31 months. 415

The results shown here suggest that the dripwater transfer time at all monitored drip sites in Bunker Cave ranges between 29 to 31 months (ca. 2.5 years). This is in good agreement with tritium data, suggesting a transfer time of 2-4 years (±1 year; Kluge et al., 2010). The tritium-based estimates, however, represent mixing ages of younger infiltrating water and older water of the epikarst reservoir (Kluge et al., 2010; Riechelmann et al., 2012a). 421 Based on the fact that all dripwaters plot on the LMWL, alteration of  $\delta^{18}$ O and  $\delta$ D values are 422 excluded and hence, both retain the temperature signal of rainwater.

Summing up, the multi-annual monitoring programme at Bunker Cave sheds light on
the complex processes affecting water transfer times. Obviously, a short-term monitoring
program would fail to resolve these patterns.

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#### 427 5.3 Influence of the North Atlantic Oscillation on the climate in western Germany

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429 The North Atlantic Oscillation (NAO) regulates Northern Hemisphere climate variability and particularly so in western Europe and eastern North America (e.g., Marshall et 430 al., 2001; Visbeck et al., 2001; Baldini et al., 2008; Hurrell and Deser, 2010; Langebroek et 431 al., 2011; Pinto and Raible, 2012; Trouet et al., 2012; Wassenburg et al., 2013, 2016; Comas-432 Bru et al., 2013). The NAO index is defined as the difference of sea-level pressure between 433 434 the Icelandic Low and the Azores High (Hurrell, 1995; Wanner et al., 2001). While the sealevel pressure pattern is present throughout the year, it is most pronounced during winter 435 (Wanner et al., 2001; Pinto and Raible, 2012). Thus, most often the winter (December to 436 March) NAO index is used when studying climatic variability in the North Atlantic realm 437 (Proctor et al., 2000; Trouet et al., 2009; Baker et al., 2015). During a negative NAO mode, 438 the winter in Europe is dominated by cold and dry conditions, while during a positive NAO 439 mode, mild and humid conditions prevail (Fig. 11A; Hurrell and van Loon, 1997; Marshall et 440 al., 2001). This NAO-temperature link is reflected in  $\delta^{18}$ O of precipitation (Baldini et al., 441 2008; Mischel et al., 2015; Comas-Bru et al., 2016) and thus can be reconstructed using 442 suitably sensitive speleothems. 443

444 To quantify the effect of the NAO on regional weather and climate, precipitation and 445 temperature records of the GNIP stations Bad Salzuflen, Emmerich, Koblenz, Wasserkuppe

Rhön, and the meteorological stations Hagen-Fley and Hemer were averaged for the months 446 December to March. December-March average NAO index values (NAO<sub>DIFM</sub>) were 447 calculated using data provided by the Koninklijk Nederlands Meteorologisch Instituut 448 (KNMI; http://www.knmi.nl/). Both precipitation amount ( $r \ge 0.36$ ,  $p \le 0.05$ ,  $n \ge 19$ , except 449 Hemer) and temperature ( $r \ge 0.58$ ,  $p \le 0.03$ ,  $n \ge 5$ ; except Wasserkuppe/Rhön) correlate 450 significantly with the NAO<sub>DIFM</sub> for most stations (Fig. 11). Thus, precipitation amount and 451 temperature above Bunker Cave are affected by the NAO during winter months. Since both 452 parameters are imprinted on drip rate and the dripwater isotope composition, it is possible to 453 reconstruct NAO variability using proxy time-series from speleothems of Bunker Cave. As 454 the precipitation in this region is equally distributed throughout the year, substantial summer 455 precipitation between April to November - despite evapotranspiration - may weaken the 456 imprint of the winter NAO signal on speleothems (Mischel et al., 2015). However, studies by 457 458 Baldini et al. (2008) and Comas-Bru et al. (2016) show that many Central European regions are sensitive to reconstruct NAO variability. A moderate connection between  $\delta^{18}$ O and NAO 459 variability has also been detected for Herbstlabyrinth-Adventshöhle in Central Germany 460 (Mischel et al., 2015), supporting the notion that speleothems from these caves record NAO 461 variability. 462

With short residence times of <3 years and limited signal smoothing in the overlying epikarst, Bunker Cave proves to be a sensitive site for high-resolution proxy reconstructions of past climate dynamics under the influence of North Atlantic atmospheric circulation. The links between Bunker Cave hydrology and the NAO found in this study are consistent with the interpretation brought forward by previous workers (Fohlmeister et al., 2012; Wassenburg et al., 2016).

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470 5.4 Wider implications of the present data set for speleothem research in general

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In case of Bunker Cave, Mg/Ca ratios as well as  $\delta^{13}$ C values depend on drip rate and thus, on precipitation and infiltration amount (Riechelmann et al., 2011, 2013; Fohlmeister et al., 2012). Therefore, both proxies are well-suited for reconstructions of precipitation and NAO variability.

Disentangling air temperature based on speleothem  $\delta^{18}$ O is a more complex task. 476 There is a clear temperature signal in dripwater  $\delta^{18}$ O of all drip sites. This temperature signal 477 of speleothem  $\delta^{18}$ O, however, is altered by (drip rate-related) fractionation processes during 478 calcite precipitation in the cave (Riechelmann et al., 2013). In the case of speleothem  $\delta^{18}$ O in 479 Bunker Cave, variations in both parameters (precipitation and temperature) are recorded 480 481 (Fohlmeister et al., 2012). A proxy not recorded in speleothem calcite but in dripwater is δD showing a well-defined temperature signal. Fluid inclusions extracted from speleothems could 482 be used to quantify  $\delta D$  and  $\delta^{18}O$  of the original dripwater and to constrain past temperature 483 changes (McGarry et al., 2004; van Breukelen et al., 2008; Affolter et al., 2015). Given that 484 the temperature pattern is unaltered in dripwater  $\delta^{18}$ O, fluid inclusions  $\delta^{18}$ O and  $\delta$ D analyses 485 486 should allow for the reconstruction of past temperature variability from speleothems in Bunker Cave and thus, variations of NAO strength. 487

The most striking observation of the present study is the difference between hydraulic response time, i.e. immediate drip rate response (<1 year; Fig. 7), and the transfer time of water from the soil to the cave (ca. 2.5 years; Fig. 10). While drip rate response to annual precipitation occurs without significant lag on (sub-)annual basis, the temperature signal is delayed by up to 2.5 years. Thus, the precipitation and temperature signals recorded by different proxies in speleothems are not fully synchronous. In case of Bunker Cave this delay is negligible, because here speleothems record proxies on decadal rather than annual or 495 seasonal scales (Riechelmann et al., 2011; Fohlmeister et al., 2012). Such proxy biases to
496 either water transfer or hydraulic response time must be considered in (sub-)annual
497 reconstructions, and where the signal transfer times for both parameters differ.

498

499 **6.** Conclusions

500

The seven year-long monitoring of hydrological parameters in and above Bunker Cave, NW Germany, provides unprecedented insights into processes influencing  $\delta^{18}$ O and  $\delta$ D in dripwater and speleothems that cannot be resolved with shorter-term monitoring programmes. The results of this study aim at quantifying the most relevant processes that may alter isotope proxies in Bunker Cave speleothems over time scales of several years:

Inter-annual drip rate dynamics are controlled by annual infiltration and thus, annual
 precipitation. No lag of the hydraulic response time is observed at annual resolution.

508 2. Water transfer time is 3 months for a 40 cm-thick soil layer and 4 months for a 70 cm-thick
509 soil. It takes approximately 2.5 years for meteoric water to reach the cave chambers.

510 3. Neither oxygen nor hydrogen isotopes of dripwaters are altered and hence, record the 511 temperature signal of meteoric precipitation. As a consequence,  $\delta^{18}$ O and  $\delta$ D from fluid 512 inclusions in speleothems are genuine temperature proxies at this site.

4. Temperature and precipitation during winter months (December-March) are significantly
influenced by the NAO. Hence, long-term NAO variations are recorded in speleothems
from northwestern Germany.

5. Monitoring data reveal a difference between the hydraulic response time and the water
transfer time. This, however, does not affect the proxy interpretation of speleothems from
this cave, because of their slow growth rate. But, with regard to other caves, especially

- with seasonally to annually resolved speleothems, this time offset may lead to difficultiesregarding the interpretation of various proxy data.
- 521 6. This study provides insights into multi-annual processes operating in cave depositional
  522 environments that are of relevance in speleothem research and can serve as a template for
  523 other cave monitoring programmes.
- 524

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526

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- 783

#### 784 Figure Captions

785

Fig. 1 Geological map of the northern Rhenish Slate Mountains in northwestern Germany.
Locations of Bunker Cave, German Cave Museum Iserlohn and the meteorological stations
(MS) Hagen-Fley and Hemer are shown (modified after Riechelmann et al., 2011).

789

Fig. 2 Plan view of Bunker Cave with the locations of drip sites TS 1 and TS 5 in chamber 1
and TS 2, TS 3 and TS 8 in chamber 2 (modified after Scheudoschi and Hasenkamp in
Scheudoschi, 2007).

793

Fig. 3 Monthly values of temperature as well as rainwater oxygen and hydrogen isotope data 794 and mean annual values of precipitation, infiltration, temperature as well as rainwater oxygen 795 and hydrogen isotope data. A) Precipitation data from 2000 to 2006 of the meteorological 796 797 station (MS) Hagen-Fley and data from 2007 to 2013 measured during this study at the German Cave Museum Iserlohn. Temperature data from 2000 to 2007 were provided by the 798 MS Hagen-Fley and 2008 to 2013 by the MS Hemer. Infiltration data are calculated using 799 data of both meteorological stations. B) Rainwater oxygen and hydrogen isotope data 800 measured from the rain sampled at the German Cave Museum Iserlohn. 801

802

Fig. 4 A) Comparison of mean annual oxygen isotope data of four GNIP stations with
rainwater data collected during this study. B) Comparison of mean annual hydrogen isotope
data of four GNIP stations with rainwater data collected in this study. C) Local meteoric water
line (LMWL) of the rainwater and position of the dripwaters on the LMWL (small box).

807

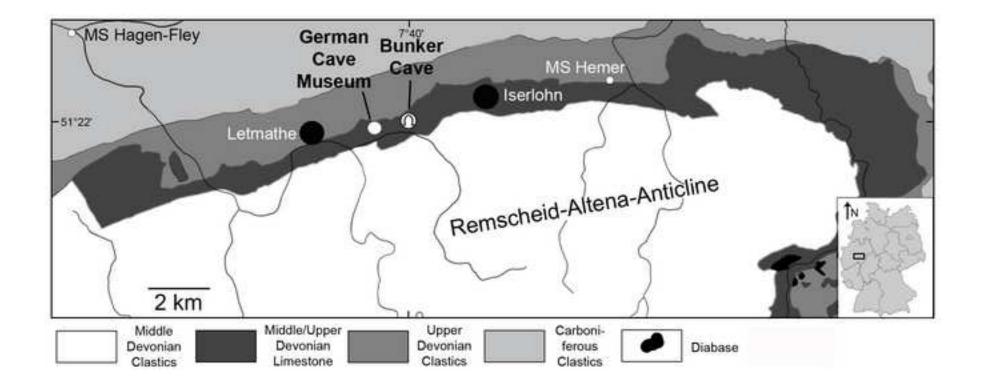
Fig. 5 Mean annual drip rates in Bunker Cave. Both manual and automatic drip rate data areshown.

810

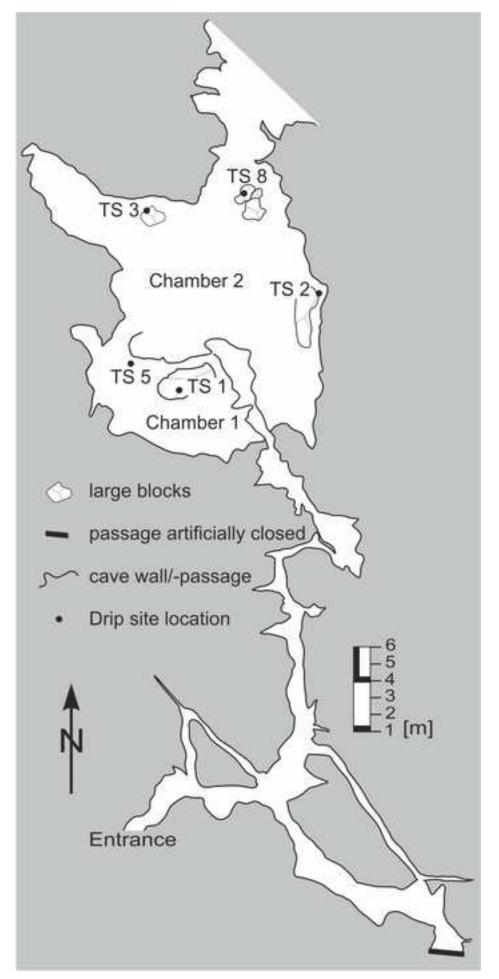
Fig. 6 Monthly and mean annual oxygen and hydrogen isotope data of soil- and dripwaters.

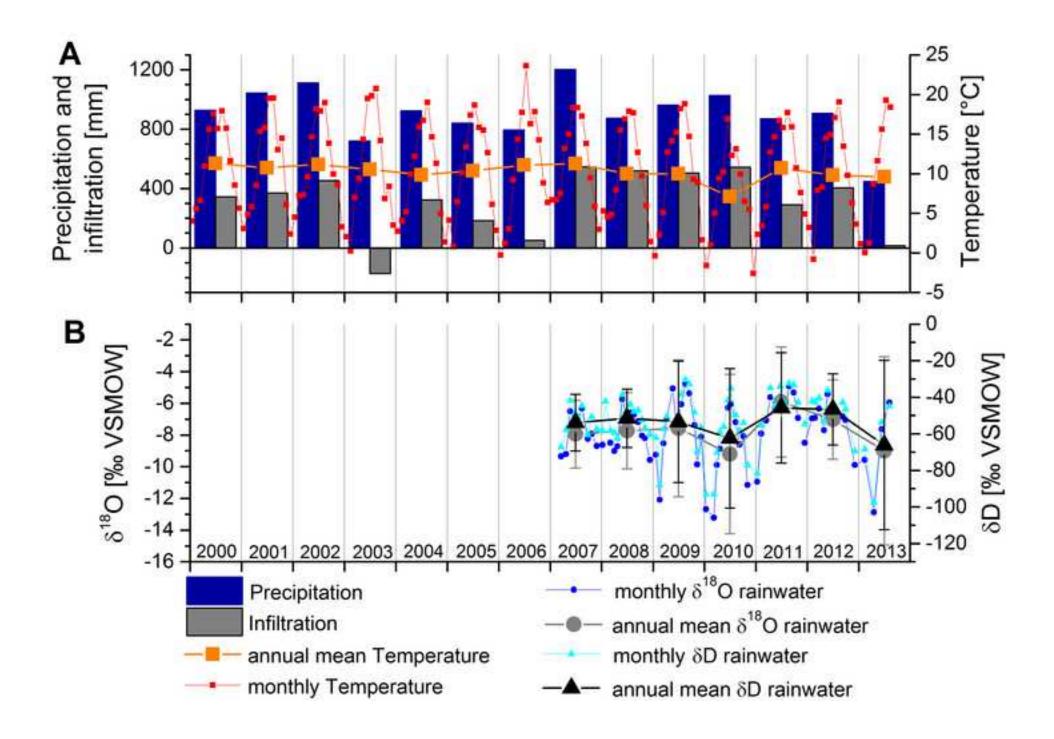
813	Fig. 7 A) Correlation coefficients between mean annual drip rate and annual infiltration. B)
814	Correlation coefficients of mean annual drip rate with annual precipitation. Note the offset to
815	obtain the hydraulic response time of the reservoir to precipitation and infiltration.
816	
817	Fig. 8 Drip characteristics of drip site TS 8 according to Smart and Friederich (1987) and
818	Baker et al. (1997). Data from 2013 are not displayed as they do not cover the whole year.
819	
820	Fig. 9 Correlation coefficients of monthly $\delta^{18}O$ and $\delta D$ values with atmospheric temperature
821	for both soilwater sites BW 1 and BW 2.
822	
823	Fig. 10 Correlation coefficients between the monthly $\delta^{18}O$ and $\delta D$ values of all dripwaters and
824	atmospheric temperature.
825	
826	Fig. 11 A) NAO index for December to March calculated for the period 1975-2013
827	(http://www.knmi.nl/). B) Precipitation data of the GNIP and meteorological stations for

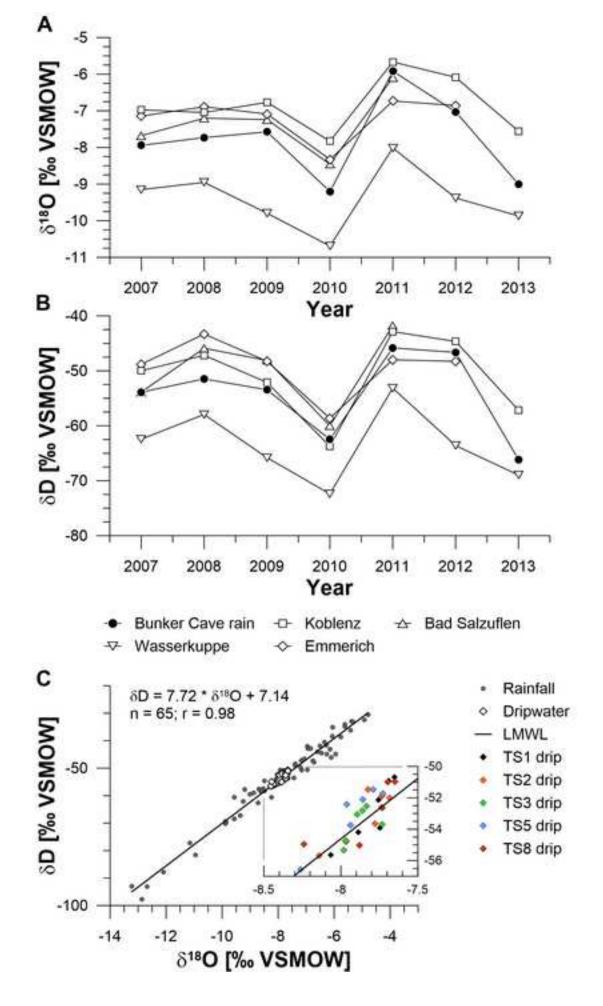
B28 December to March for the period 1975-2013. C) Temperature data of the GNIP andmeteorological stations for December to March for the period 1978-2013.

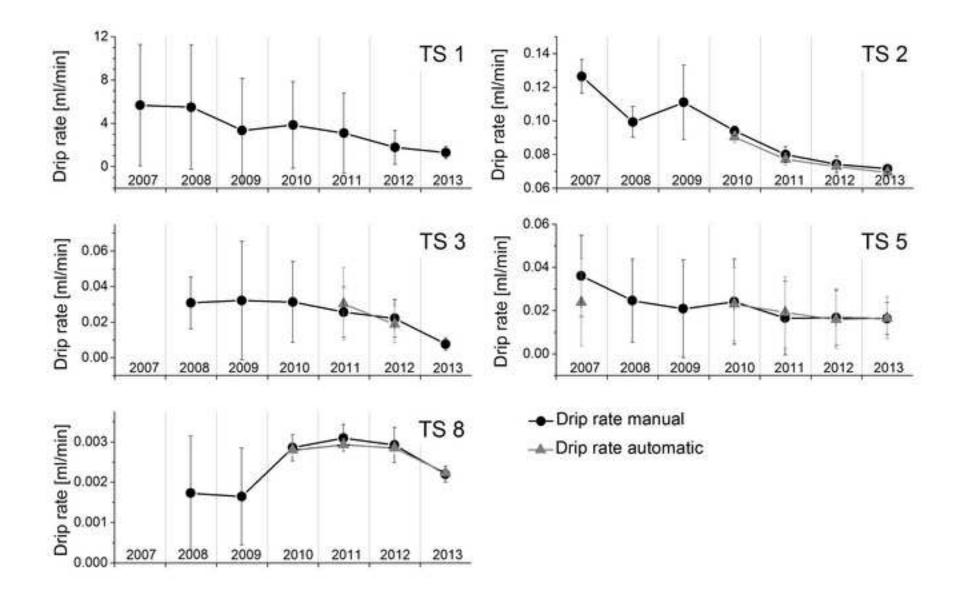


## Figure 2 Click here to download high resolution image









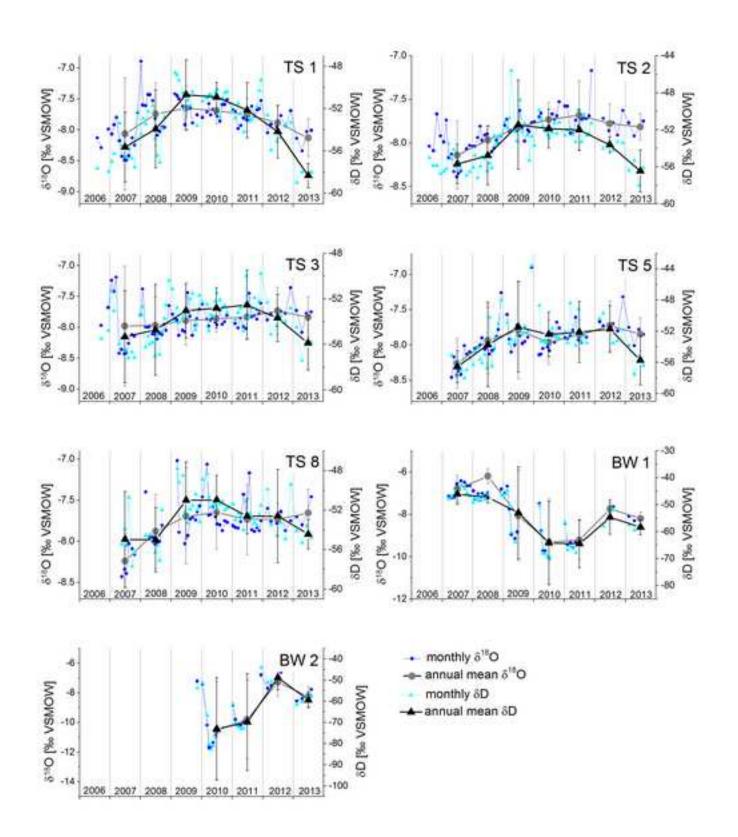
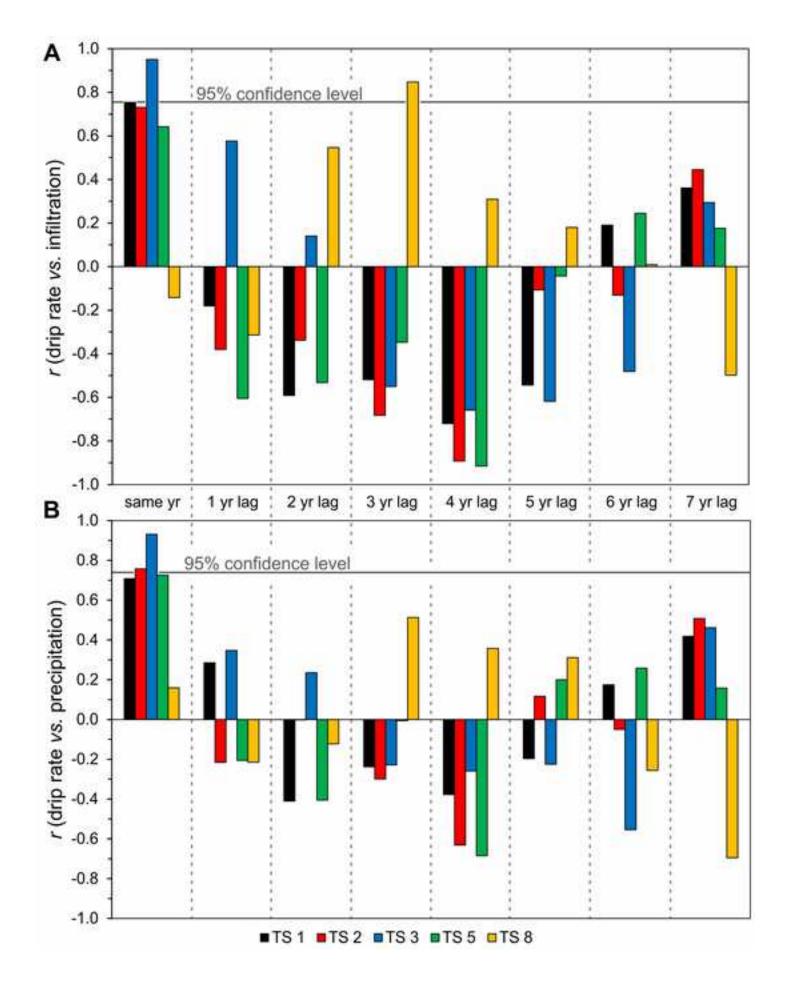
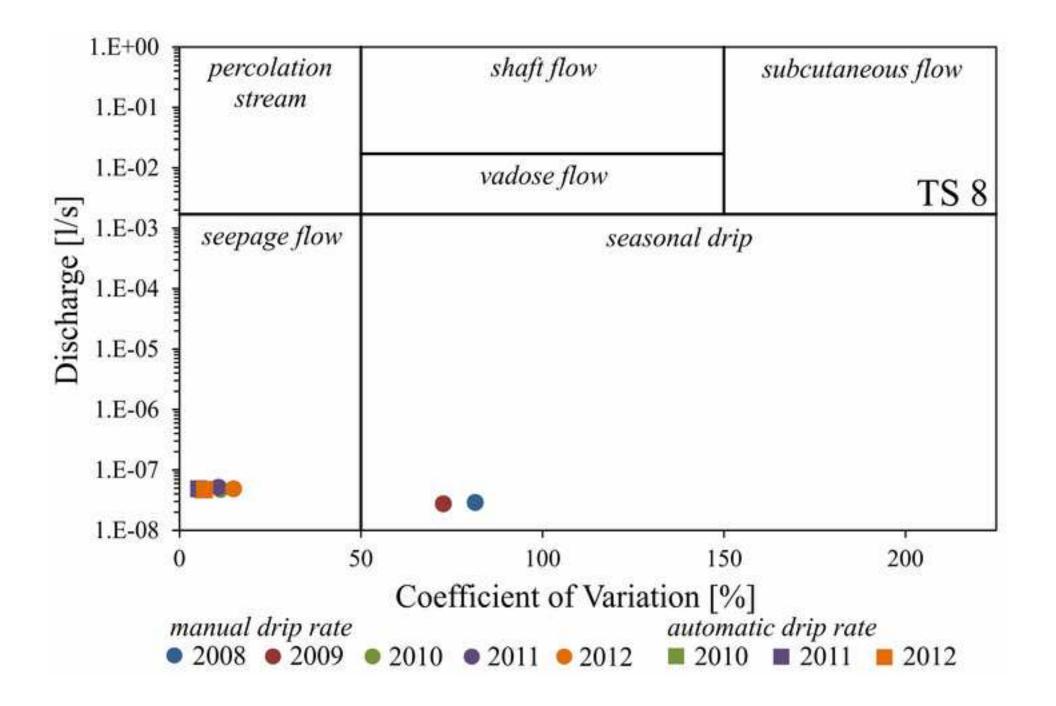
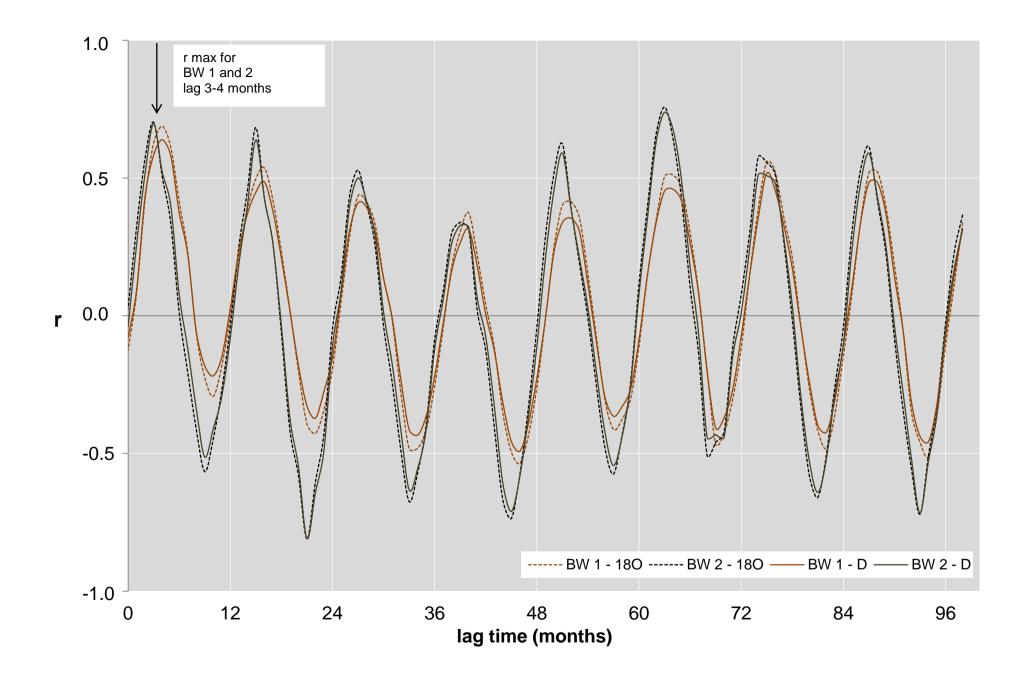
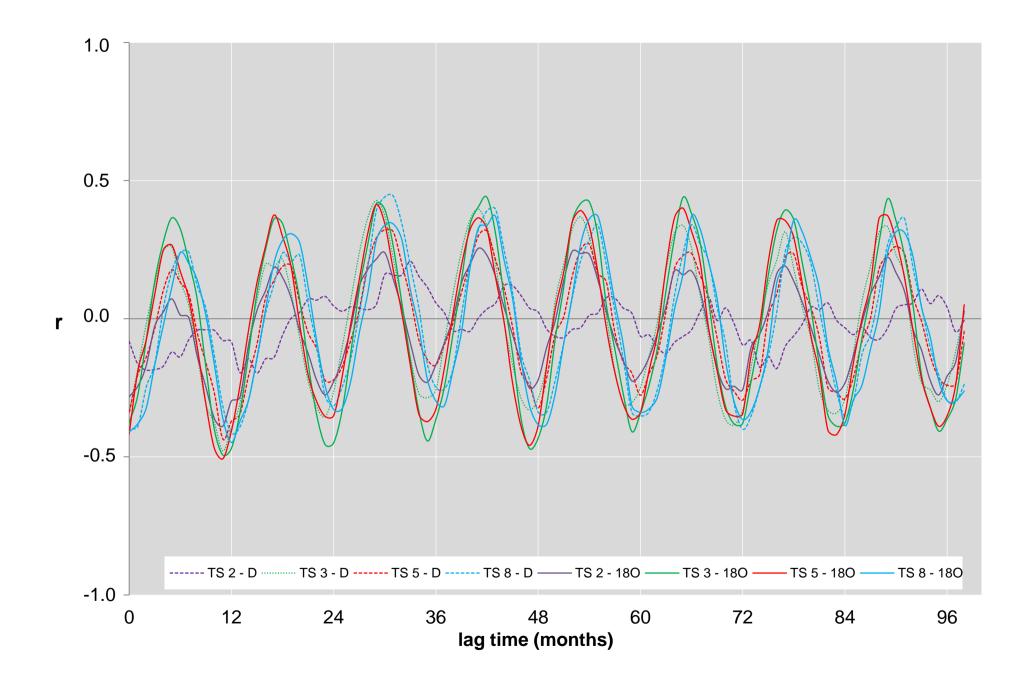


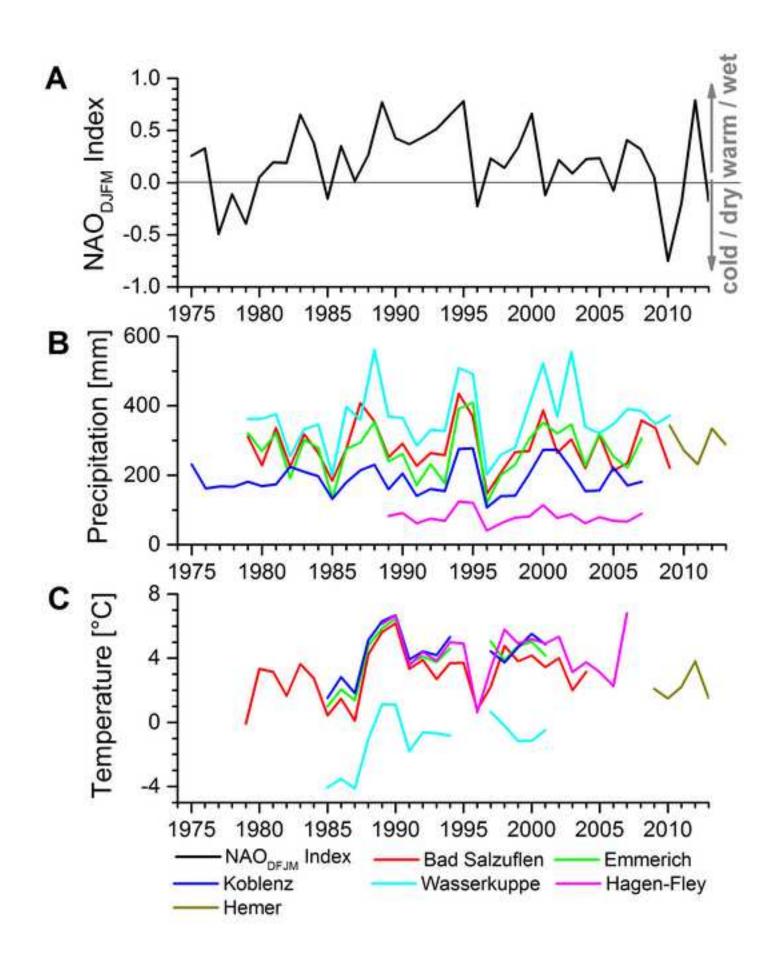
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**BW** 1

BW 2

-0.11

-0.27

-0.53

-0.05

, mineant (		coefficients	(r, p = 0.0)	o) are print		itu pointo t
tistics var	y between 14	4 and 65.				
δ <sup>18</sup> 0	TS 1	TS 2	TS 3	TS 5	TS 8	BW 1
TS 2	0.53	10 2	10.0	100	10.0	<b>D</b> (( 1
TS 3	0.36	0.43				
TS 5	0.41	0.55	0.63			
TS 8	0.50	0.47	0.58	0.37		

-0.07

0.57

-0.17

0.66

-0.31

-0.07

0.72

Table 1. Correlations between monthly  $\delta^{18}$ O values of the different drip- and soilwaters. Significant correlation coefficients (r;  $p \le 0.05$ ) are printed bold. Data points used for statistics vary between 14 and 65.

Table 2. Correlations between monthly $\delta D$ values of the different drip- and soilwaters.	
Significant correlation coefficients (r; $p \le 0.05$ ) are printed bold. Data points used for	
statistics vary between 15 and 65.	

δD	TS 1	TS 2	TS 3	TS 5	TS 8	BW 1
TS 2	0.74					
TS 3	0.61	0.59				
TS 5	0.51	0.55	0.73			
TS 8	0.72	0.63	0.61	0.45		
BW 1	-0.34	-0.45	-0.38	-0.37	-0.38	
BW 2	-0.34	-0.58	0.07	0.04	-0.14	0.76

Background dataset for online publication only Click here to download Background dataset for online publication only: Supplementary material.docx