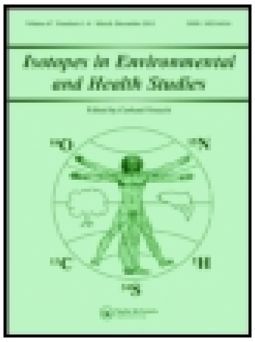

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Estimation of groundwater recharge via deuterium labelling in the semi-arid Cuvelai-Etosha Basin, Namibia[†]

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The stable water isotope deuterium (^{2}H) was applied as an artificial tracer ($^{2}H_2O$) in order to estimate groundwater recharge through the unsaturated zone and describe soil water movement in a semi-arid region of northern central Namibia. A particular focus of this study was to assess the spatiotemporal persistence of the tracer when applied in the field on a small scale under extreme climatic conditions and to propose a method to obtain estimates of recharge in data-scarce regions. At two natural sites that differ in vegetation cover, soil and geology, 500 ml of a 70 % $^{2}H_2O$ solution was irrigated onto water saturated plots. The displacement of the ^{2}H peak was analyzed 1 and 10 days after an artificial rain event of 20 mm as well as after the rainy season. Results show that it is possible to apply the peak displacement method for the estimation of groundwater recharge rates in semi-arid environments via deuterium labelling. Potential recharge for the rainy season 2013/2014 was calculated as 45 mm a^{-1} at 5.6 m depth and 40 mm a^{-1} at 0.9 m depth at the two studied sites, respectively. Under saturated conditions, the artificial rain events moved 2.1 and 0.5 m downwards, respectively. The tracer at the deep sand site (site 1) was found after the rainy season at 5.6 m depth, corresponding to a displacement of 3.2 m. This equals in an average travel velocity of 2.8 cm d^{-1} during the rainy season at the first site. At the second location, the tracer peak was discovered at 0.9 m depth; displacement was found to be only 0.4 m equalling an average movement of 0.2 cm d^{-1} through the unsaturated zone due to an underlying calcrete formation. Tracer recovery after one rainy season was found to be as low as 3.6 % at site 1 and 1.9 % at site 2. With an *in situ* measuring technique, a three-dimensional distribution of ^{2}H after the rainy season could be measured and visualized. This study comprises the first application of the peak displacement method using a deuterium labelling technique for the estimation of groundwater recharge in semi-arid regions. Deuterium proved to be a suitable tracer for studies within the soil–vegetation–atmosphere interface. The results of this study are relevant for the design of labelling experiments in the unsaturated zone of dry areas using $^{2}H_2O$ as a tracer and obtaining estimations of groundwater recharge on a local scale. The presented methodology is particularly beneficial in data-scarce environments, where recharge pathways and mechanisms are poorly understood.

Keywords: deuterium labelling; groundwater recharge; hydrogen-2; peak displacement; semi-arid areas; unsaturated zone

1. Introduction

Over large parts of semi-arid Sub-Saharan Africa (SSA), where population growth rates are high and industrialization is progressing, pressure on water resources is steadily increasing.

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Groundwater resources are therefore becoming progressively more important as they can act as a secure source of freshwater, for example, during periods of drought or during the dry season. In order to manage aquifers sustainably, an accurate determination of recharge rates is crucial. Groundwater recharge is widely acknowledged as being the most important parameter for the management of aquifers [1]. At the same time, it is the parameter that is most difficult to determine [2]. Consequently, many researchers are developing methods for the quantification of groundwater recharge. Tracer-based methods are amongst the most precise methods for groundwater recharge studies [3,4]. However, quantitative studies on groundwater recharge using stable isotope methods are difficult and rather rare [4,5].

The peak displacement method [6,7] has been proven to be a suitable technique for a quantitative estimation of groundwater recharge. Even though processes such as dispersion and preferential flow are difficult to tackle, the method is a simple approach to obtain estimations of recharge. The technique has been successfully applied in areas where a significant seasonal variation of the concentration of the stable isotopes deuterium (^{2}H) and oxygen-18 (^{18}O) in precipitation is present and is reflected in the soil water stable isotope profiles [6,8]. Numerous precedent studies utilized the peak displacement method using tritium (^{3}H) in the past. Tritium was imprinted into the rain and subsequently soil water by thermonuclear bomb testing, and successfully used to derive groundwater recharge rates in semi-arid regions [5,9–14]. A complete review is given by Koeniger et al. [15]. Furthermore, tritium has been used as an artificial tracer in numerous studies in various climatic regions using different experimental procedures [16–24].

Within the last decade, only a few studies using the peak displacement method, using either environmental isotopes or artificial labeling, have been conducted due to several reasons.

Even though a seasonal variation of isotopic composition of ^{2}H and ^{18}O might be detectable in the rain, this signal is often lost within the soil profile due to dispersion effects and mixing [3,25]. An application of the method using environmental tracers, in particular in semi-arid regions, is therefore often not possible.

In the southern hemisphere in particular, the impact of the thermonuclear tests was much less pronounced than in the northern hemisphere. Tritium concentrations have returned to close to environmental concentrations and are hence not practically useful for this method anymore.

1. (1) Within the unsaturated zone, the peak concentrations resulting from the thermonuclear tests can only be found in greater depths (>20 m). Hence, sampling and analysis of soil profiles become very labour intensive and no longer feasible.
2. (2) Tritium is toxic if used in high concentrations and therefore of concern to both the environment and health. This limits its practicability.
3. (3) Transient flow and transport models are now available to account for the whole spectrum of involved processes (e.g. dispersion, preferential flow) and can be applied where detailed information on soil hydraulic properties is available.

Recently, artificial applications of the stable isotope deuterium are becoming increasingly popular in the fields of hydrology and ecohydrology. Labelling with ^2H_2O is of advantage because it is not radioactive and there are no toxicological concerns during both labelling and measurement [26]. Being part of the water molecule, ^{2}H is considered a conservative tracer and thus suitable for studies in the unsaturated zone [27]. It can be measured in very low concentrations [26]; therefore only small amounts of tracer are necessary and applications become economically feasible.

In numerous studies, ^2H_2O is applied as an artificial tracer to investigate water movement in the unsaturated zone. In their pioneer efforts, Zimmermann et al. [16,17] used deuterium for the study of soil water movement. They found in their experiments that in sandy soils water transport occurs layered, that is, if new water infiltrates into a soil, the old water is simply pushed downward [16]. They investigated dispersion effects and quantified recharge in a humid region in Sweden over a period of several months based on peak displacement. A comparison of deuterium, bromide and chloride during an irrigation experiment to assess the uncertainty of tracer experiments under similar boundary conditions was presented by Lischeid et al. [28]. An investigation of preferential flow through a forest-reclaimed lignitic mine soil was conducted by Hangen et al. [29]. Results showed that deuterium and bromide showed similar transport behaviour and were considered as suitable tracers for studying preferential flow. Similarly, Schumann and Herrmann [30] identified preferential flow paths during an irrigation experiment. Mali et al. [31] studied water movement through the unsaturated zone using ^2H_2O and dye tracer. The authors were able to quantify travel velocities and vertical dispersion and concluded that deuterium is a useful tool for unsaturated zone studies and more suitable than dye tracer for such applications. Koeniger et al. [27] applied ^2H, ^{18}O and uranine in column and field studies. They estimated flow rates through the unsaturated zone and compared the tracers. Mean transport velocities for deuterium were found to be higher and dispersion coefficients lower compared to uranine in the column experiments. The authors confirmed ^2H_2O to be a suitable water tracer extending possibilities for field studies in the field of biogeosciences. Recently, Grünberger [32] was able to quantify capillary rise from a shallow aquifer using deuterium as an artificial tracer.

As shown, many precedent studies deal with water movement in the unsaturated zone. Studies focusing on the quantification of recharge via labelling with ^2H_2O are rare, in particular within the unsaturated zone of semi-arid regions. The fate of deuterium applied as an artificial tracer within the unsaturated zone of soils in semi-arid climates over a longer period (e.g. one rainy season) has not been investigated. Such climates are characterized by high evaporation rates, the presence of a dry season, heterogeneous soils and specialized vegetation. In addition, groundwater tables are generally far from the surface. This imposes additional challenges for both experimental design and theoretical considerations of recharge studies. The potential of using ^2H_2O for the purpose of quantifying deep percolation and estimating recharge in such environments has not been assessed yet.

In fact, there is currently no established method for the quantification of groundwater recharge in semi-arid climates based on stable isotopes of water. The persistence of artificially introduced ^2H_2O in the unsaturated zone of such areas is yet to be studied, and the necessary input concentrations and methods of application are poorly understood. Becker and Coplen [26] explicitly state that ‘further research is required to better understand how isotopic exchange affects the behavior of deuterated water over the time scale of a tracer experiment’. Additionally, it is in (semi-)arid areas where the portfolio of available methods for recharge estimation is further limited due to scarcity of data, lack of infrastructure or high spatiotemporal variability.

The main objective of this study is to adapt the method developed by Saxena [33] for the direct quantification of potential annual groundwater recharge through the unsaturated zone in the Cuvelai-Etosha Basin (CEB) using artificial deuterium labelling. We aim to investigate water movement in the unsaturated zone and fate of ^2H_2O introduced by an artificial rain event over the course of an entire rainy season. A further focus is set on challenges regarding the experimental design of such experiments under extreme climatic conditions.

2. Study area

The CEB is a transboundary endorheic watershed shared almost equally by Angola and Namibia with a total size of 173,000 km^2. A digital elevation model of the CEB is presented in Figure 1. The Cuvelai-Etosha region is home to a large number of people both on the Namibian and Angolan sides, mainly because of the presence of shallow groundwater and relatively fertile

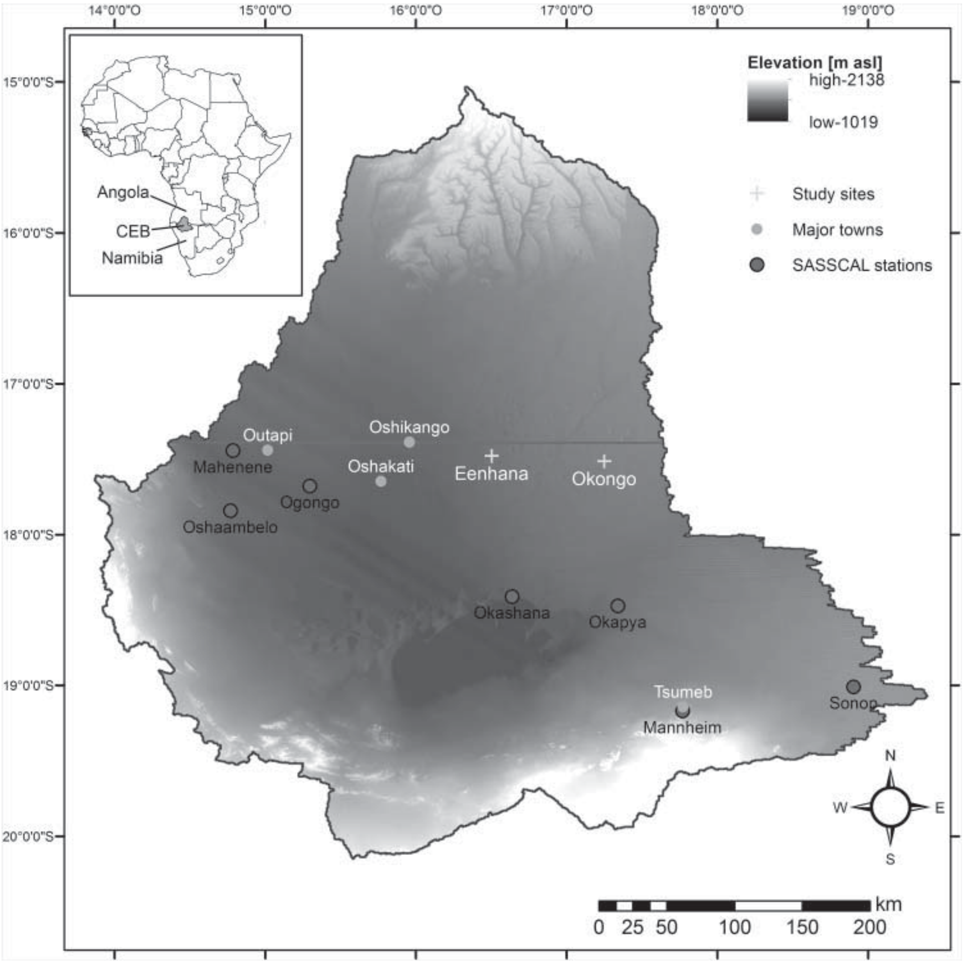

Figure 1. Digital elevation model of the transboundary Cuvelai-Etosha surface water Basin (CEB). Marked are the locations of major towns, climate stations established in the framework of South African Science and Service Center for Climate Change, Agriculture and Landuse (SASSCAL) and the study sites for the presented research. The grey line marks the borders of Namibia and Angola.

soils in many areas [34]. The basin has a vivid hydrogeological history as both the deltas of the Cunene and Okavango Rivers were once situated within the CEB [35,36]. In recent history, a deep aquifer containing fresh water was discovered in the northeastern part of the basin [35]. At present all of the surface water is either draining towards the Etosha Pan, a salt pan in the southern part of the CEB, or remains in surface depressions (locally called *iishana*) that are forming a vast, partly inter-connected channel-like system north-west of the Etosha Pan [34]. No perennial river exists, and the basin receives all of its water concentrated over the rainy season from November to April. Mean annual precipitation varies between 200 and 600 mm a^{-1} along a distinct rainfall gradient from the west to the east of the basin [34]. Temperature average throughout the basin is higher than 22 °C with maximum values reaching up to 40 °C in summer [34]. In winter, temperatures can drop to around zero at night. Evaporation rates can reach up to 3000 mm a^{-1} and exceed yearly rainfall by a factor of five. In Figure 2, long-term rainfall records from the station Ondangwa and rainfall distribution of 2013/2014 at Elundu school, Eenhana constituency, are shown. Groundwater recharge processes are poorly understood due to the complex discharge

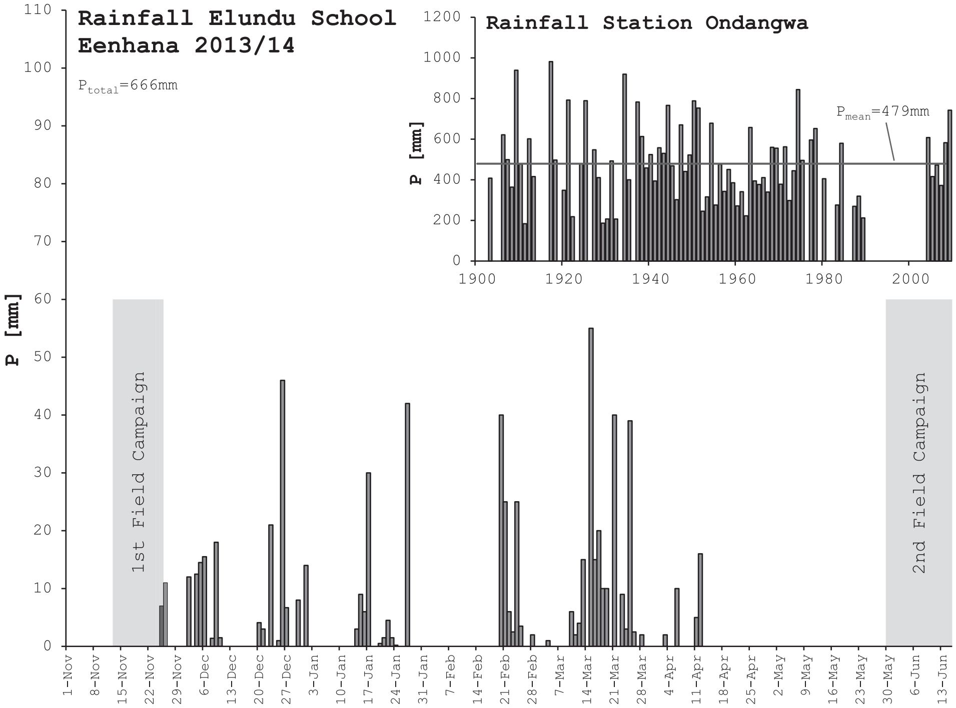

Figure 2. Rainfall for the rainy season 2013/2014 measured at Elundu school, Eenhana and long-term rainfall at the station Ondangwa (inset graphic). Note that gaps in the long-term series at Ondangwa represent missing years.

and rainfall patterns and the high spatial variability of soils, vegetation and geology. *Iishana*, ephemeral river beds, a high number of pans, dunes and calcrete formations create direct and indirect recharge paths which until now have been difficult to describe.

Two sites differing in soil type, geological set-up and vegetation within the CEB were chosen for this study: (1) Eenhana – a forested site on deep Kalahari sand and (2) Okongo – a site with woodland and shrubs on loamy sand underlain by a thick calcrete layer. The selected study sites represent major land forms throughout the CEB. At both sites, soils are classified as arenosols [34]. The groundwater table at these two sites is 30 and 20 m below the surface, respectively. Around the Eenhana site (site 1), the pure sand can reach depths of several hundred metres, and deep infiltration of rain water can be expected. Resulting from visual inspection and preliminary irrigation experiments, no hydrophobic behaviour of the soils was observed. At the top layer (first 3 cm), a biological soil crust was identified which likely developed due to extreme temperatures and dryness during the Namibian winter in combination with microbiological activity. Within the first 20 cm below surface, finger-like flow was observed. The soil is homogeneous and, according to its soil hydraulic properties, highly permeable (Table 1). At the Okongo site (site 2), none of these effects was observed. Soils in this area contained generally more silt and clay, and hydraulic conductivity is lower than at site 1 (Table 1). The hydraulic behaviour of the thick (often more than 10 m) calcrete layer is unclear. Double-ring infiltrometer tests on top of the calcrete showed no immediate infiltration of water. After using a well camera at several hand-dug wells on-site, cracks and macropores potentially enhancing recharge became visible. Information on soils and local vegetation are summarized in Table 1. The latter aspect plays an important role within the hydrological cycle of (semi-)arid environments as it was recognized as a main factor controlling deep drainage and groundwater recharge [37]. A conceptual description of common rooting strategies in such regions is

Table 1. Rainfall, vegetation and soil characteristics as well as parameters determined in the laboratory for experiments conducted at the Eenhana and Okongo sites in the CEB, Namibia.

		Eenhana	Okongo
Total rainfall 2013/2014	(mm a^{-1})	660	660[a]
Vegetation type	–	Forest	Woodland
Dominant plant species	–	*Combretum collinum*; *Acacia erioloba*; *Baikiea plurijuga*	*Collinum apiculatum/zeyheri*; *Ricinodendron rautenii*
Maximum rooting depth	(m)	2.3–2.4	1.0–2.0
Soil type			
0–100 cm	–	Sand	Loamy sand
Sand/silt/clay	(%)	98/1/1	94/3/3
$\gg$100 cm	–	Sand	Calcrete
Sand/silt/clay	(%)	97/2/1	79/7/14
Bulk density	(g cm^{-3})	1.59	1.53
Porosity	–	0.35	0.40
Field capacity (pF = 2.5)	(vol.-%)	3.5	5.1
Residual water content	(vol.-%)	3.3	4.5
Saturated water content	(vol.-%)	35.3	39.5
Saturated hydraulic conductivity (lab)	(cm d^{-1})	2490	1184
Saturated hydraulic conductivity (field)	(cm d^{-1})	2304	1872
Size of plot	(m^2)	0.27	0.27
Depth of ^2H_2O injection	(m)	0.25	0.25
Amount of tracer irrigated	(ml)	500	500
Amount of tracer irrigated	(mm)	1.85	1.85
Depth of tracer peak before rainy season	(m)	2.3	0.4
Depth of tracer peak after rainy season	(m)	5.5	0.9
Groundwater recharge	(mm a^{-1})	45	41
Percent of precipitation	(%)	~7	~6

[a] Approximated. Distance between Eenhana rain gauge and Okongo study site is about 100 km.

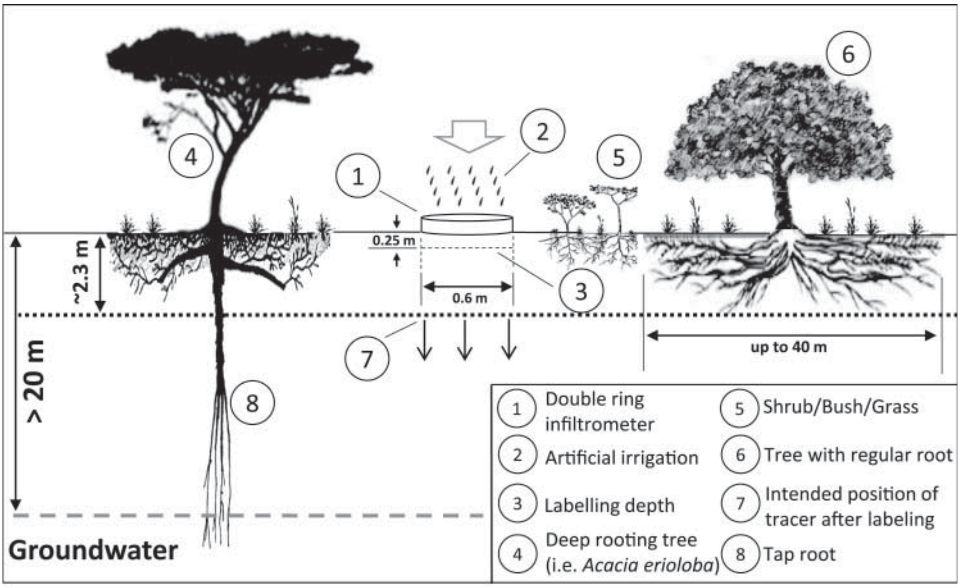

Figure 3. Experimental design and typical characteristics of vegetation in semi-arid and arid environments.

incorporated in Figure 3. The tree species present throughout the study area (e.g. *A. erioloba, B. plurijuga*, *C. collinum*; Table 1) are known to be capable of adaptations to particularly dry conditions, such as the development of deep tap roots [38,39]. We refer to the discussion section for a comprehensive analysis of the impact of local vegetation on the genesis of groundwater recharge.

3. Methods

3.1. *Experimental design*

For the design of the experiments, special attention has to be paid towards the amount and method of tracer application. The main objective for this research is to inject the ^2H_2O before the start of the rainy season and detect it during the subsequent dry season. The two major requirements to achieve this are (i) to prevent the tracer from evaporating and (ii) not to lose the entire tracer solution via root water uptake through plants. As a consequence, the tracer needs to be either injected or moved to a depth where root activity is at minimum or at least markedly reduced and transpiration is close to zero. This is of great importance to assure that the tracer signal would be detectable after the rainy season. Diffusion effects are of minor importance when looking at purely sandy soil (compare soil hydraulic properties in Table 1) during the rainy season since vertical water transport is dominating. To fulfill the above mentioned prerequisites, an experimental procedure was developed as follows.

First, typical rainy season conditions (wet soil) within the experimental plot comprising an area of 0.27 m^2 were established and saturated conductivity (*ks*) derived by performing a double-ring infiltrometer test. After gentle excavation of the sand until the target depth of 25 cm, 1.85 mm (500 ml) of 70 % ^2H_2O was distributed uniformly using drip irrigation with an average intensity of 0.75 mm m^{-1}. Finally, after carefully refilling the soil, an artificial rain event of 20 mm was irrigated onto the plot with a mean intensity of 2.5 mm min^{-1}. To examine the position of the tracer front, one soil profile each with a vertical sampling resolution of 10 cm was taken using an Eijkelkamp hand auger 1, 2 and 10 days after the application of ^2H_2O until a maximum depth of 2.6 m (site 1). At the second site, profiles were taken after 1, 2 and 5 days. After the rainy season, samples were taken up to a maximum depth of 7.4 m. The soil was collected in headspace glass vials that were sealed with a crimping tool immediately after sampling. From these samples, the isotopic composition, water content and grain size distribution were determined in the laboratory (refer to laboratory methods). Additional plant samples were taken at site 2 since during the course of the field experiment, fresh grass was growing directly on the experimental plot and similarly analyzed for isotope ratios.

A recent development allowed the measurement of stable isotopes in soils directly in the field and was helpful to decide on the maximum sampling depth. Even though this method is still in an experimental stage, the approximate magnitude of ^2H values gave an idea as to whether there was a tracer in the samples and at which depth the values would return to the background concentration. For this *in situ* measurement of ^2H and ^{18}O, commercially available soil gas probes (BGL-30, Umweltmesssysteme (UMS), Munich) were connected to a laser spectrometer (OA-ICOS, Los Gatos Research, DLT100). A laptop communicating with the OA-ICOS was programmed to control the measurement cycle and to mimic the behaviour of a laboratory auto sampler. In essence, the step involving the vapourization of a liquid sample was skipped and the soil gas directly sent to the measuring cavity of the OA-ICOS. Measurements of stable isotopes were accordingly taken at a resolution of 30 cm horizontally and 20 cm vertically starting from the centre of the experimental plot. This resulted in 22 field measurements that were interpolated using the statistic software R [40,41]. The procedure enabled creating a highly resolute image of the deuterium distribution above the calcrete layer and a quasi-3D visualization of the spatial ^2H distribution after the rainy season at site 2.

A schematic description of the experimental design is presented in Figure 3.

3.2. *Laboratory analysis*

Gravimetric water content was determined after drying the samples for 24 h at 105 °C. The grain size distribution was derived by dynamic image analysis. For fine soils the grain size distribution smaller than 63 μm was analyzed by sieving and sedimentation [42]. Soil hydraulic properties, that is, water retention parameters and saturated as well as unsaturated conductivities, were obtained using the evaporation method [43–45] and subsequent fitting of retention curves with the software ROSETTA [46]. Saturated conductivity was also examined in the field using a double-ring infiltrometer. In Table 1, the most important parameters describing the soil hydraulic behaviour of both soils are compiled.

For the extraction of soil water, a cryogenic vacuum distillation was used, which is described in detail by West et al. [47] and was modified for higher throughput by Koeniger et al. [48]. Despite recent developments of measuring stable isotopes *in situ* [49,50], cryogenic vacuum extraction still is the most commonly used method and applied to extract water from soil, xylem and plant samples [29,47,48]. In the present study, two replicate samples for each depth with approximately 10 g of soil were prepared and vacuum extracted at 105 °C. Even for the driest samples, a minimum of 0.1 ml of water could be extracted from those.

The extracted water samples were analyzed for ^{2}H concentrations using a Picarro L2120-i cavity ring-down (CRD) water vapour analyser after vapourisation, and can be expressed in the unit [$10^3\ \delta^2$H] following the definition of Coplen [51] given in Equation (1):

$$10^3\delta^2\mathrm{H} = \left(\frac{R_{\mathrm{Sample}}}{R_{\mathrm{Standard}}} - 1\right), \tag{1}$$

with R_{Sample} being the ratio (^{2}H/^{1}H) of the less abundant to the more abundant isotope in the sample and R_{Standard} the ratio (^{2}H/^{1}H) in a standard. For high concentrations it is advantageous to express the concentrations in parts per million (ppm), as given in Kendall and McDonnell [52]:

$$^2\mathrm{H}\ (\mathrm{ppm}) = \left(\frac{\delta^2\mathrm{H}}{1000} + 1\right)^{*}156 \tag{2}$$

Subsequently, this notation will be used throughout the manuscript.

3.3. *Estimation of local groundwater recharge and tracer recovery*

In the 1960s, a method was developed to quantify groundwater recharge rates in sandy soils by tracing natural ^{18}O and injected tritium profiles [16,17]. Saxena and Dressie [6] define the downward movement of soil moisture as the shift of the peak concentration of the tracer in the case of artificial labelling, and as the mean depth of the depleted isotope layer when looking at the seasonal variability of stable isotopes. The rate of soil moisture movement was calculated using Equation (3):

$$v = \frac{z_2 - z_1}{t_2 - t_1}, \tag{3}$$

where v [L T^{-1}] is the velocity of soil moisture movement from the tracer depth (as defined above) z_1 to z_2 [L] during the time span between t_1 and t_2 [T]. The amount of water stored within the soil S [L] during this period is then simply calculated by multiplying the soil moisture θ [–] with the vertical extent of the particular soil layer:

$$S = \theta \cdot \Delta z. \tag{4}$$

Assuming the tracer peak is located underneath the root zone and water surpassing the root zone is not subject to further effects, recharge through the unsaturated zone R_{un} [LT^{-1}] can be determined as follows:

$$R_{\text{un}} = \frac{\int_{z_0}^{z_n} \theta(z) \cdot dz}{T}, \tag{5}$$

where z_0 and z_n [L] represent the water depths of the tracer peak after injection of the tracer and after a certain time period T (i.e. one rainy season). In our study, we follow the above-described approach with two adaptations to the methodology:

1. Since there is no rain during the dry season, the influence of summer rain can be neglected [6]. Hence recharge can be calculated from the position of the tracer peak before (t_1) and after (t_2) the rainy season.
2. In order to avoid the overestimation of the calculated recharge due to the initial saturated conditions, a correction of water contents was introduced. We use as a threshold water content at pF 2.5 (i.e. the lower end of field capacity) and define any water with a lower pF as draining water. From the clearly visible peak in water content caused by the artificially introduced water, the difference between field capacity and observed water content is calculated for each soil layer (Equation (6)):

$$I_{\text{corr}} = (\theta_{\text{obs}} - \theta_{fc}) \cdot \Delta z, \tag{6}$$

where I_{corr} [L] represents the water depth that is to be corrected for, θ_{obs} is the observed water content and θ_{fc} the water content at field capacity for the particular soil layer. By implementing Equation (6) into Equation (5), the final equation for the estimation of recharge can be expressed as:

$$R_{\text{un}} = \frac{\int_{z_0}^{z_n} \theta(z) \cdot dz}{T} - \int_{z_b}^{z_e} I_{\text{corr}} \cdot dz, \tag{7}$$

where z_b and z_e [L] represent the depths of the beginning and the end of the artificially introduced event, respectively.

The calculated recharge rate represents the amount of water infiltrating underneath the lateral root zone. We subsequently refer to this as ‘potential recharge’ because of the possibility that this water is subject to effects (e.g. water vapour transport) during the dry season.

When introducing artificial water into a soil, the movement of subsequent rain events might occur at an accelerated rate due to the relationship of hydraulic conductivity and water content (i.e. higher water content in the soil profile leads to higher hydraulic conductivity). At both study sites, however, the upper 30 cm of the soil profile dried out within less than three days. In semi-arid areas, the typical evapotranspiration rate is 5–8 mm d^{-1} [52]. Therefore, the hydraulic connectivity between the artificially introduced water and the rain is lost very quickly. A major rain event would then be needed to re-establish this connection. Hence, for the presented study, the impact of the artificially introduced water is reduced to a minimum. Keeping track of the soil water balance in such places can help determine the impact of the artificially introduced water.

To assess the persistence of $^2\text{H}_2\text{O}$ in the unsaturated zone, tracer recovery was calculated according to the procedure described by Speakman [53]. An assumption had to be made that only the volume underneath the irrigated plot was considered, that is, lateral movement of the tracer was neglected. Homogeneity of ^{2}H concentrations was assumed for the complete soil layer where the sample was taken.

4. Results

4.1. *Characterization of water movement distribution of ^2H_2O at Eenhana forest site*

Volumetric water content profiles 1 and 10 days after tracer injection as well as after the rainy season are presented in Figure 4(a). Downward movement of the introduced 'package' of water can be clearly observed. One day after the injection of ^2H_2O, the event water has already infiltrated to a depth of more than 1 m. In the subsequent days, excess water travels slower through the drier media below. For the period between the application of the tracer and the 10-day profile, the infiltration rate equals 23 cm d^{-1}. Measurements after the rainy season indicate that the soil moisture is constantly below field capacity from the soil surface until about 2.3 m below the surface, which reveals the effect of evapotranspiration. Notably, a clear increasing step-shift of moisture content at 2.4 m is observed, which is due to the end of the root zone. This has been further investigated and proved by both visual inspection of the depth profiles and further tracer experiments. Underneath 2.4 m the sand at the investigated site has an almost white colour with no visual signs of organic contamination. The average travel velocity over the whole rainy season resulting from the after rainy season profile is 2.8 cm d^{-1}. At a depth of around 6–6.5 m, the water content drops to almost dry marking the end of moisture front of the rainy season 2013/2014. The elevated soil moisture contents near the surface 1 and 10 days after the irrigation are caused by overnight rain events.

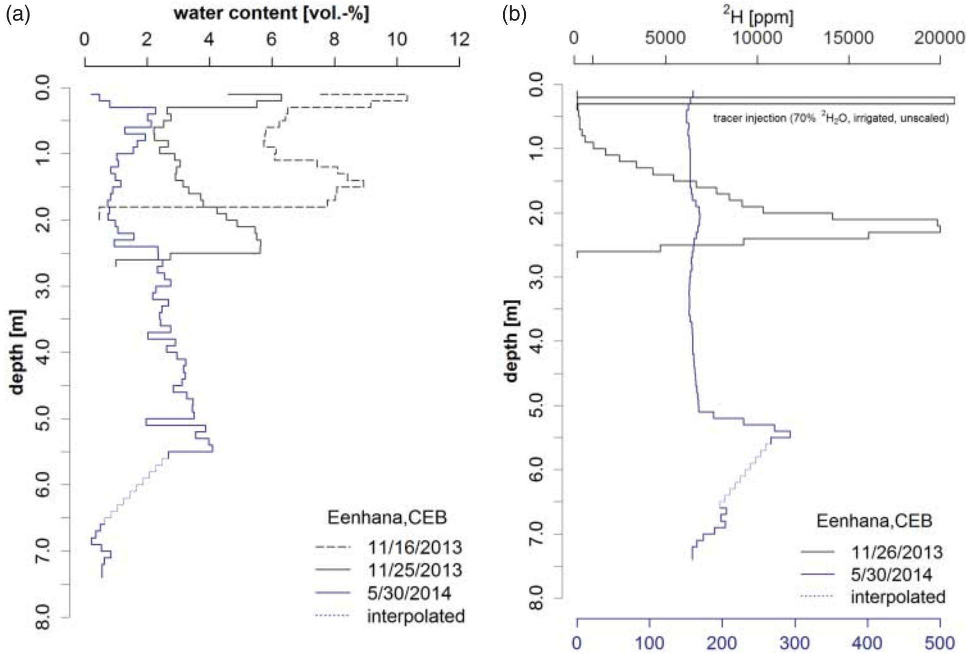

Figure 4. (a) Water content profiles during the experiment for the Eenhana site. (b) Deuterium concentrations during the experiment for the Eenhana site. Note that two different axes are used for ^{2}H concentrations for better visualization. The upper x axis belongs to the profile prior to the rainy season (26 November 2013) and the lower x axis belongs to the after rainy season profile (30 May 2014).

The ^2H profiles prior to and after the rainy season (Figure 4(b)) provide a clear picture of the displacement of soil moisture during the rainy season. A Dirac-like type of pulse of the tracer injection is reflected within the soil profile as a sharp peak in the 10-day profile. Advection (downward displacement) as well as dispersion (tailing of the tracer front) effects can be observed in the post rainy season profile. In the upper 5 m below ground, the tracer concentrations remain constant and at similar concentrations as the non-tracer-influenced parts of the shallow (pre rainy season) profiles with two exceptions: (i) the after rainy season profile (Figure 4(b)) also shows that the artificially introduced ^2H_2O must have remained at that depth for a certain time. An elevated deuterium concentration can still be identified at that depth, which might indicate an exchange with immobile water; and (ii) a clear evaporation effect is visible in the upper 0.3 m. The peak tracer concentration after the rainy season (~ 300 ppm) at 5.5 m is reduced by a factor of 70 compared to the peak concentration of the 10-day profile (~ 21,000 ppm). The after rainy season profile shows that precipitation during the rainy season shifted the peak downwards to 5.5 m below ground. This corresponds to a peak displacement of 3.2 m. At 7.4 m depth, the deuterium concentration moves back to background concentration (158 ppm) marking the end of the tracer front. It was found that 10 days after the tracer application, 26.2 % of the input was still present within the soil. After the rainy season, the number of ^2H molecules present in the soil profile equals only 3.6 % of the input.

4.2. *Characterization of water movement distribution of ^2H_2O at Okongo shrub–woodland site*

At the second study site, a significantly different infiltration behaviour of the moisture front was observed (Figure 5(a)). One day after the tracer injection, significant peaks in soil moisture are present at ~ 0.3 and 0.7 m. A further increase of water content due to impoundment

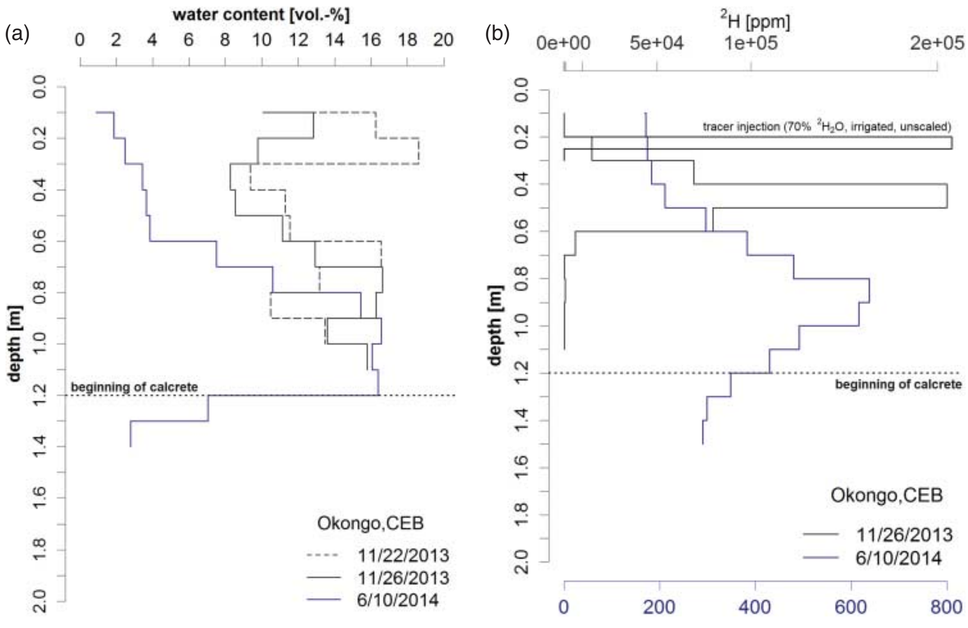

Figure 5. (a) Water content profiles during the experiment for the Okongo site. (b) Deuterium concentrations during the experiment for Okongo. Note that two different axes are used for ^2H concentrations for better visualization. The upper x axis belongs to the profile prior to the rainy season (26 November 2013) and the lower x axis belongs to the after rainy season profile (10 June 2014).

is pronounced towards the beginning of the calcrete layer. The majority of the infiltrated water is, however, found between 0.3 and 0.4 m depth. This indicates that water movement at the Okongo site is much slower and processes are different from those at the Eenhana site. Five days after labelling, the (now homogenized) moisture front introduced is clearly visible at a depth of approximately 0.9 m. For this period, infiltration velocity equals 0.13 cm d^{-1}, which is almost half of that at the Eenhana site. This is in accordance with observed differences in soil hydraulic characteristics between the two sites (Table 1). Similar to changes in water content, deuterium transport was restricted to the upper part of the profile. Peak concentrations of deuterium five days after the tracer injection are found at 0.5 m. The tracer peak after the rainy season is found at 0.9 m depth. This corresponds to a peak shift of only 0.4 m or an average travel velocity of 0.2 cm d^{-1}, being much smaller than those for the Eenhana site. Tracer recovery was calculated from the post rainy season profile, and it was found that 1.9 % of the tracer applied remained within the soil underneath the experimental plot. No infiltration of water underneath the root zone was observed, which is due to the calcretic layer starting at 1.2 m depth hampering deeper sampling. The water content on top of this layer is significantly above field capacity, and the impounding water is expected to evaporate, be taken up by roots, or infiltrate into the groundwater via potentially existing preferential flow paths along plant roots or cracks and fissures within the calcrete layer. These results imply consequences for the application of the peak displacement method at this site as discussed below. The recovery rate of the tracer was calculated as 1.6 % after the rainy season.

4.3. *Spatial distribution of the tracer*

To examine the spatial distribution of the remaining tracer after the rainy season, Figure 6 presents an x–z and x–y plot of the distribution of ^2H at Okongo. It shows a quasi-3D visualization of the spatial ^2H distribution. The following insights for the presented experiment as well as for the further application of ^2H_2O tracer in the unsaturated zone can be derived:

(1) Over the course of the rainy season, a significant portion of the applied tracer was retained in the soil matrix. This fact is surprising since one would expect a loss of the tracer in hot climates, where evaporation and transpiration loss is high and plants are flourishing during the rainy season. This demonstrates that 500 ml of 70 % ^2H_2O was a sufficient amount of tracer for this study.

(2) Diffusion and mixing effects distributed the tracer over a much larger area than the initial irrigated plot which was 60 cm in diameter. Horizontally, ^2H concentrations greater than 300 ppm can still be found more than 60 cm from the centre of the plot. In the upward vertical direction, the influence of the tracer reaches up to $\sim$20 cm below the surface. An upward transport of the tracer by evaporation is the most probable reason for this.

(3) Highest tracer concentrations were found at 90 cm depth directly underneath the area of tracer application. Maximum concentrations of 500–550 ppm in x direction coincide with the dimensions of the experimental plot. In the z direction these maximum concentrations were found over a smaller length (25 cm), which is most likely due to a non-uniform spread of the tracer during application or heterogeneities in the soil matrix such as root channels or locally occurring calcrete accumulations.

(4) The centre of mass of ^2H_2O did not reach the calcrete layer. This is possibly caused by a slight change of soil hydraulic properties towards the pure calcrete and is dependent on the genesis of the calcrete layer. Lateral movement of water at this depth might become more dominant. However, this cannot be further evaluated due to missing *in situ* measurements below 60 cm.

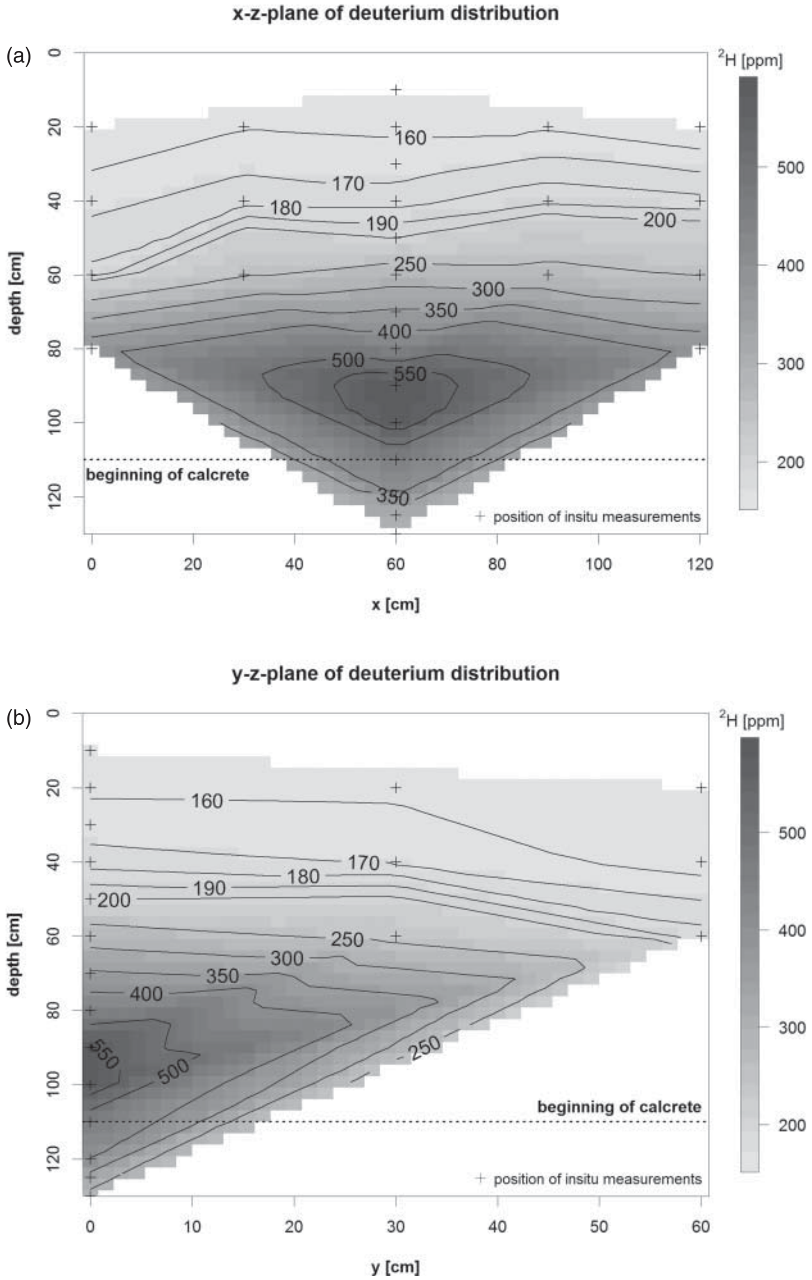

Figure 6. (a) Spatial distribution of ^2H_2O after the rainy season at the Okongo site in x–z direction. (b) Spatial distribution of ^2H_2O after the rainy season at Okongo site in y–z direction.

4.4. *Calculation of groundwater recharge*

The calculation of potential groundwater recharge rates follows the methodologies described above (Section 3). At the Eenhana site, a correction for the artificially introduced water was necessary due to the fact that the increased soil moisture starting at 1.7 m depth (Figure 4(a)) is expected to cause infiltration underneath the root zone. Hence, this portion of water had to be accounted for. After this correction, the direct groundwater recharge rates through the unsaturated zone were calculated as 45 mm for the rainy season 2013/2014 at this site corresponding to 7 % of the rainfall. According to these results, the deep sands around Eenhana comprise a potential recharge area of the CEB.

At Okongo, a potential recharge of 40 mm was estimated. However, due to the impermeable layer at around 1.2 m, the tracer peak had to be applied within the root zone. We use the term 'potential recharge' in this case since the majority of this water is still available for plants and potentially exposed to evaporation during the dry season. Explicitly stated, it is possible that practically none of this water might reach the groundwater table. Table 1 is summarizing the results for both studied sites.

5. Discussion

5.1. *Soil water movement and distribution of ^2H_2O*

The presented profiles of soil water movement and tracer movement enable a site-specific characterization of the unsaturated zone water movement. With the combination of tracer and soil moisture profiles, quantification of water fluxes is possible. Especially at Eenhana, we emphasize the importance of the intra-seasonal distribution of rainfall for the movement of water through the unsaturated zone and the creation of groundwater recharge in semi-arid environments, which is discussed in earlier studies [54]. From further experiments on-site, it was determined that a rainfall event as high as 60 mm did not infiltrate deeper than 40 cm on a dry soil (data not shown herein). Looking at the soil hydraulic properties of the soil at the Eenhana site (Table 1), it becomes clear how low the water-holding capacity of the media is. For longer wet periods and extreme events, infiltration of water underneath the root zone can be expected. It is therefore crucial for recharge estimates to investigate the rainfall characteristics of the rainy season when evaluating groundwater recharge. An analysis of these characteristics according to the criteria defined in Beyer et al. [54] was performed for the rainy season 2013/2014. Indicators which are believed to be related to groundwater recharge such as maximum wet spell duration, number of events above 10 mm and rainfall during the core rainy season [54] were found to be in an upper range: rainfall within the core rainy season was 615 mm, and the maximum wet spell duration equalled 22 days. Furthermore, 19 events exceeding 10 mm were recorded (rainfall events >10 mm are commonly considered as productive rainfall events, [34]). It is these events that create conditions for deep infiltration of water [52]. Only four events reached magnitudes greater than 40 mm. These results imply that in this particular rainy season, longer wet periods rather than extreme rain events caused the deep infiltration of water and thus recharge. For further research this points out the potential of such field experiments which can be combined with transient flow and transport modelling and analysis of rainfall distribution within the rainy season. In addition to soil moisture, including tracer transport and distribution into model calibration will lead to more robust models and could potentially be used for developing local empirical relationships for the estimation of groundwater recharge [3].

The after rainy season profile at Eenhana shows how soil moisture increases with depth. In this case, it was possible to identify the end of the lateral root zone at around 2.3–2.4 m depth. However, the role of deep-rooting tree species, which are utilizing a tap root (compare Figure 2) to support the tree with moisture during the dry season, has to be further investigated. Such tree species (e.g. *A. erioloba*, *B. plurijuga*) can potentially reach groundwater tables as deep as 60 m [55,56]. This has to be accounted for when quantifying groundwater recharge. To our knowledge, little is known on the volume of water 'pumped' from the groundwater table by these region-adapted plant species. In addition, it is not clarified until now if such roots are able to utilize water in greater depths of the unsaturated zone. This issue could be tackled by injecting ^2H_2O into the root of such trees and quantify dry season transpiration using the tracer concentration breakthrough curve during future research activities.

Whereas at Eenhana the interpretation of the combined ^2H_2O and soil moisture profiles provided clear evidence, difficulties arise when interpreting the results for the Okongo site. From the soil moisture profiles one and five days after injection of ^2H_2O, respectively, one could infer that the zone of main root activity is probably located between a depth of 0.3 and 0.5 m. The after rainy season profile reveals an impoundment of water at the calcretic formation. Below this layer, water content decreases with depth, which might be due to either very little infiltration or a decrease in porosity. Comparing the peak of the soil moisture with the one of ^2H, it becomes obvious that these measurements do not coincide. Only a slightly elevated concentration of deuterium is found at the position of maximum soil moisture. This could be interpreted as follows: the main amount of water was introduced artificially by the preliminary water saturation of the soils. This ‘unlabelled’ water represents the moisture front in the five-day profile. The actual tracer application and subsequent artificial rain event only reached a depth of 0.5 m with a small portion of the tracer reaching the pre-saturation-induced moisture front via preferential flow paths causing the small increase of ^2H_2O at 0.9 m. Another indicator for the presence of preferential flow is the difference in field and laboratory determined saturated hydraulic conductivity ks (see Table 1). In addition, we used a dye tracer and visually inspected its distribution after several irrigation experiments. Preferential flow within the upper 50 cm could clearly be identified. However, another explanation might be that ‘old’ water simply is pushed downwards.

5.2. *Tracer recovery*

With 3.6 % at Eenhana and 1.9 % at the Okongo site, tracer recovery determined after one rainy season was very low at both sites. Nevertheless, the peak concentrations of ^2H were clearly detectable (Figures 3 and 4). Loss of tracer can be attributed to the combined effects of evaporation, transpiration and lateral flow. At Eenhana, almost 75 % of the applied tracer disappeared already 10 days after the application. We believe this is caused mainly by evapotranspiration. The root zone at the Eenhana site extends deeper than at Okongo. If irrigating the tracer, it first has to surpass the root zone and is therefore subject to transpiration. Furthermore, the experiments took place during the flowering season, where transpiration demand is very high. Upward transport could be verified by sampling and analysis of fresh grass growing on top of the experimental plot and xylem samples of shrubs nearby at Okongo. The results show that all grass samples which were taken two and five days after the application of ^2H_2O showed considerably elevated concentrations (>200 ppm) of ^2H indicating an upward transport of the tracer to the surface (the background concentration is ~155 ppm). Samples of shrubs approximately 1 m from the plot that were taken two days after the experiment showed no enrichment (155 ppm). A slightly elevated concentration of ^2H (~160 ppm) was found in a grass sample outside of, but close to, the experimental plot indicating lateral transport of the tracer. The fact that at both studied sites the tracer concentration was well within the detectable range confirms the suitability of ^2H_2O for experiments under semi-arid conditions.

5.3. *Uncertainty and shortcomings of recharge estimates using peak displacement*

The groundwater recharge rates determined in this study at both sites agree well with the range of values that were found in previous studies (between 20 and 100 mm, [57]). It should be noted that the determined recharge is a direct recharge for a year with exceptionally high rainfall at the study site. Therefore, the value of 45 mm a^{-1} should rather be seen as an upper percentile and does perhaps not represent the mean recharge which might be lower. In addition, it has to be kept in mind that the determined recharge through the unsaturated zone does not necessarily coincide with recharge at the groundwater table. This is only the case if no other effects such as water vapour transport from greater depths or uptake of deep soil water through tap roots of specialized plants are present. The presented methodology, although being the most direct way of quantifying recharge, is subject to a certain degree of uncertainty due to a number of factors:

(1) Heterogeneity and the contribution and potential effect of preferential flow must be acknowledged. If sampling in the field occurs along a preferential pathway, this will result in an overestimation of groundwater recharge because the tracer would penetrate much deeper. Similarly, if preferential pathways are present, but the sampling does not occur along it, recharge would be underestimated. This issue has been pointed out by several authors [23,58]. A possible way of overcoming this problem might be to compare the results with estimations based on the chloride mass balance method [1,59]. Sampling multiple profiles in the field within the experimental plot, labelling at more than one plot per site and an identification of lateral flow, might further help distinguish between preferential and matrix flow. Hence, the uncertainty of the method can be lowered by taking into account local heterogeneities.

(2) Any irrigation is introducing artificial water into the soil column. Hence, any water possibly contributing to infiltration has to be accounted for. Furthermore, following the principles of soil sciences, soil hydraulic conductivity is increasing with a higher water content potentially causing a change of initial conditions for the rainy season. In the example of Eenhana, this turned out to be of minor consequence because in the 10 days after the injection of the deuterium, 51 mm or 5.6 mm d^{-1} was lost within the soil profile due to evapotranspiration. Soil hydraulic connectivity between the artificial rain event and the top soil was cut, that is, the top soil already had dried out up to a depth of 1.5 m (Figure 4(a)). However, for future research a different experimental design not using irrigation should be considered. Currently, a punctual application of deuterium in depth is tested causing no disturbance of the experimental plot and applying only a minimum amount of water.

Since the development of the peak displacement method [6,16], no considerable methodological advances have been reported to account for non-mobile water or heterogeneities. In the original technique [6], the water content between the position of the tracer peak after application and after a certain time is used to calculate recharge. But in fact, not all water within a soil profile is mobile, that is, there is always a residual portion of water which is not contributing to recharge per se. If the soil hydraulic properties of the site under investigation can be determined precisely, the residual water content should be subtracted and included in Equation (5).

Very little research has been conducted on the uncertainty of the method. In existing studies [22,60–62], uncertainty for the method ranges in between 1 and 20 %. Considering applied laboratory and field methodologies of the presented study, our estimation is confidently within that range. A challenge is that even if water is infiltrating underneath the primary root zone, it could potentially be lost through upward water vapour transport during the dry season. In addition, the role of deep groundwater table penetrating roots should be investigated further as this comprises a potential sink component of the groundwater balance.

Finally, uncertainty of the method could be introduced by the set-up and calibration of transient flow and transport models. These types of model are becoming increasingly popular and have been applied successfully in the past [8,63]. Advantages of such models are the ability to incorporate effects such as dispersion, preferential flow or water vapour transport. Several researchers are working on the development of models which are able to simulate fractionation processes of stable isotopes of water [64–66]. This will contribute to improve process understanding, in particular in (semi-)arid environments. However, for data-scarce countries, the non-availability of necessary input data might limit the applicability of such detailed approaches. In addition, numerical modelling in such environments is challenging due to rapid changes in dry–wet conditions. Currently, models are being developed for the presented study sites. This might enable a quantification of local recharge over a longer period of time and investigating favourable conditions for the creation of recharge as well as improving process understanding within the unsaturated zone of semi-arid environments.

6. Conclusions

We proposed a method for a quantification of direct groundwater recharge in semi-arid areas using a single-tracer experiment following the peak displacement method. A cost-efficient and easily applicable experimental framework was used, which is delivering reliable estimations of direct recharge and allows a hydraulic characterization of the unsaturated zone.

As concluding remarks, we state the following:

(1) We presented a methodology to apply peak displacement utilizing ^2H_2O as a tracer to determine annual groundwater recharge on a local scale. Deuterium is an appropriate tracer for such applications. Hence, the portfolio of methods for estimating groundwater recharge in data-scarce environments can be extended with such field experiments. A very low amount of tracer is required (less than 500 ml) to obtain reliable results for recharge on deep soils.

(2) At sites with a near-surface impermeable layer, the methodology is difficult to apply due to the fact that at such sites, infiltration underneath the root zone does not take place. It can be expected that during the dry season this water will be utilized by the local vegetation and evaporation. Care needs to be taken when interpreting data at such sites. However, when having the opportunity to gain further insight into processes within the calcretic layers, for example, by deeper drilling, the method might still be useful.

(3) With combined moisture and tracer profiles, calibration and parameterization of transient flow and transport models can be improved in the future potentially leading to more robust models for dry climates and increase understanding of (eco-)hydrological processes in complex environments.

(4) The experimental design can be improved in future studies by punctual application of the tracer underneath the root zone. This would eliminate the issues arising with the introduction of artificial water into the soil.

(5) The role of deep-rooting plant species and preferential flow paths for the estimation of groundwater recharge needs to be investigated further, especially in dry climates where plants are adapted to local conditions. This requires interdisciplinary research (e.g. the application of ground penetrating radar or transient electro-magnetic systems). The contribution of preferential flow might be quantified by taking multiple soil cores within the experimental plot. Set-up and calibration of transient flow and transport models will help to quantify such processes. Further research should also be carried out in regard to theoretical considerations of the methodology, in particular to account for heterogeneities within the unsaturated zone. Finally, the potential effect of site-specific upward transport of water should be investigated to decrease the uncertainty of the method further and increase understanding of hydrological processes in semi-arid regions in general. ^2H_2O as a tracer, potentially combined with others, provides opportunities for future studies.

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