Author's response to the review by Christine Stumpp

We were thankful for the thoroughly review of the manuscript by Christine Stumpp. She pointed out several possible improvements that we included in the revised manuscript. In the following, we present the changes that were done for each of the points of which Christine Stumpp saw opportunities for improvements. A few points that did not lead to changes in the manuscript are discussed.

Comment:
Please emphasize that you only get information about transport (dispersivity) by using isotopes; it cannot be estimated from water content or suction data

Reply:
We emphasized this in section 1.2 as follows:

"Last but not least, pore water stable isotopes provide the means to include the transport parameter (diffusivity) into inverse modelling approaches, which would not be possible with solely water content or matric potential data."

Comment:
Why are you using deuterium contents only? What about oxygen-18? Wouldn't it add to get information about parameter uncertainty?

Reply:
We included the following paragraph at the end of section 2.1 for clarification:

"Although, the isotope analysis were done for δ^2H and $\delta^{18}O$, we only consider δ^2H in the inverse modelling approaches, because i) the relative errors of the stable isotope analysis were smaller for δ^2H with a standard deviation of 1.16‰ compared to 0.31‰ for $\delta^{18}O$, ii) ^2H is less affected by fractionation processes than ^{18}O, iii) the additional gain of information of considering both isotopes vs. just ^2H is limited, since $\delta^{18}O$ and δ^2H are highly correlated, and iv) the HYDRUS model cannot account for fractionation processes due to evaporation."

Comment:
At the three sites, the number and time point of isotope depth data are different, which has to be discussed. Having more data in the objective function (e.g. for Hartheim) most likely improves the parameter identification. Further, the model efficiency can be different for summer or winter if incorrect assumptions regarding snow melt or transpiration were made.

Reply:
We addressed the issue in the discussion at the end of section 4.1 as follows:

"The highest deviations of the modelled pore water stable isotope composition from the observed isotope profiles are found for the sampling in January and March, which could be caused by an insufficient representation of the snow melt processes or transpiration. Also

preferential flows, which were shown to occur mainly during the wet season after snow melt (Gazis and Feng, 2004; Mueller et al., 2014) might cause bigger differences between observed and simulated isotope profiles during winter times. Thus, the number of considered isotope profiles and their sampling timing can have an important impact on the inverse model approaches. Generally, it is preferable to have several pore water stable isotope profiles taken during different seasons and hydrological states.

We have seen the high influence of the transpiration on the pore water extraction via root water uptake in our simulations that followed the currently presented study. Thus, we are going to include in a future study a time variable leaf area index, estimated via site specific measurements of the solar radiation in the forest and on grassland. However, such data was not available in the current study.

Comment:

The functional evaluation procedure calculating transit times and giving annual water balance certainly adds to the interpretation of the results. With some exceptions, the annual water balance calculations are quite similar. Are they significantly different? What is the uncertainty range? The authors did a sensitivity analysis for the parameter identification. Can this be further used in the forward calculations to give ranges of the calculated water balance components?

Reply:

We extended the sensitivity analysis to the water balance calculations and transit time estimations and also apply statistics to see if the approaches result in significant different results. This new methods are described in a paragraph at the end of section 2.3:

"The sensitivities of the different approaches with regard to the water balance and transit time estimations were tested with simulations of 100 randomly chosen parameter sets from Sbest. If the different inversely determined parameter sets lead to significant different functional responses with regard to flow and transport was tested with a one-way ANOVA and a Post-hoc analysis (Tukey's HSD). The tested variables were the mean annual ET and the median transit time, defined as the time after which half of the recharge water has passed the lower boundary of the soil profile."

We describe the results for the water balance in section 3.3 as follows and list the ranges of the estimated mean transit times and the ET rates in a new Table (Table 5):

"The statistical analysis showed that the inverse model approaches resulted in significantly different mean annual ET estimates when considering the different parameter combinations of the set S_{best} (Table 5)."

and for the transit time estimations as:

"The mean transit times (MTT) simulated with 100 randomly chosen parameter combinations from S_{best}, are statistically significant different among the inverse model approaches for Eichstetten. For Roodt, transit times of the IPA and uIPA were about twice as long as for the

MOA and 2SA and the latter two approaches did not differ significantly in terms of MTT. For Hartheim, the uIPA and the MOA did not differ significantly with regard to the MTT, while the others did."

We included in Figure 5 and 6 the results of the above described procedure to visualize the results of the new sensitivity analysis. The Figure 3 is changed accordingly.

Comment:

I was expecting a more thorough discussion about the gained soil hydraulic parameters using the different approaches. Are there any data available, e.g. measured saturated hydraulic conductivity or porosities, to compare the results?

Reply:

We included comparisons between parameters derived by inverse modeling and measurements conducted on soil cores or via irrigation experiments as follows:

"As an example, measurements of Ks on soil cores taken in the catchment of the study site in Roodt showed high variability of the hydraulic conductivity with values ranging between 29 and 2306 cm day^{-1} across the soil profile. The inversely estimated Ks-values for Roodt lay within the range of these measurements. Further estimations of the MVG parameters on soil cores taken in the upper horizon in the study area at Roodt showed similar ranges as the parameter sets derived via inverse modelling. Exceptions are the parameter n, which has higher values for the uIPA and IPA than the laboratory measurements, and the θs, which is generally lower for the inverse optimization compared to the measurements, which could reflect the influence of the rock content. The deviation between the inverse estimations and laboratory measurements could also be due to the lack of high volumetric water contents in the soil moisture data and the fact that the soil moisture sensors are not calibrated. For the other study sites, no laboratory measurements on soil cores are available, but infiltration experiments with Uranin showed that that water introduced during fall events percolated down to 140 cm during one year (Koeniger, 2003) at Hartheim, which is well reproduced with the MOA and slightly overestimated by the other approaches (Table 5). Furthermore, infiltration measurements at Hartheim with a double ring revealed a high variability of the saturated hydraulic conductivity (1 – 800 cm day^{-1}) in the topsoil, and the inversely estimated Ks parameters are within this range."

Comment:

Care needs to be taken with the interpretation of dispersivities at some sites and with some approaches (see specific comments). A "spin-up" period of two years is not enough for sites with mean transit times > 2 years and therefore dispersivities in larger depths cannot be estimated.

Reply:

The spin-up period has been always at least 967 days, which is longer

We included therefore the minimum number of days in section 2.2.3 and list the number of days for every site in Table 1:

"The influence of the initial conditions on the calibration can be neglected, as a spin-up period of at least 967 days was simulated before the start of the calibration period (Table 1).

and pick up this point in the discussion in section 4.2 as follows:

"Since the efficiency of the pore water isotope simulations is beside the MVG and dispersivity parameter highly dependent on the isotopic signal of the rainwater, a sufficiently long input time series is crucial in order to ensure that the initial pore water has been renewed over the simulation period to minimize the influence of the initial conditions. In our case, this is given since the spin-up periods (Table 1) are generally longer than the estimated transit times (Fig. 6)."

Comment:
Abstract ln 16-18: it sounds like the authors did some additional HYDRUS modifications; as an existing modified version was used, it is more appropriate to write "and a modified version of HYDRUS was used allowing deuterium loss during evaporation". By the way, HYDRUS should be written with capital letter throughout the manuscript.

Reply:
We changed as suggested to:

"The transport of deuterium was simulated with the advection-dispersion equation, and a modified version of HYDRUS was used, allowing deuterium loss during evaporation."

HYDRUS is now written in capital letters throughout the manuscript.

Comment:
- 11204/11205, ln 24/ln 1, 6: "satisfying model performance", "satisfying model efficiencies": these terms are rather unspecific and subjective; re-phrase to be more specific (applies to entire manuscript)

Reply:
We changed accordingly

"Approaches b) and c) using both, the isotope profiles and the soil moisture time series resulted in good simulation results with regard to the Kling-Gupta-Efficiency and good parameter identifiability." […]Approaches b) and c) both outcompeted simulations run with parameters derived from pedotransfer functions, which did not result in an acceptable representation of the soil moisture dynamics and pore water stable isotope composition."

Comment:
- 11207, ln 8-13: referring to stream flow studies in catchments is not required

Reply:
We have included such references upon request from the editor and agree with you that the connection to the pedon scale is limited.

Comment:
- 11208, ln 3/4: "so far not been": better to write "rarely" or rewrite the sentence stating that it has not been tested how isotope depth profiles can improve the inverse modeling procedure. We previously used isotope depth profiles at four locations in Ghana to calculate groundwater recharge rates; among other methods, we also used the isotope data in HYDRUS 1D to identify flow and transport parameters (Adomako et al. 2010, HSJ, 55, 1405-1416).

Reply:
We included the reference (Adomako et al. 2010, HSJ, 55, 1405-1416) and wrote now "rarely" instead of "so far not been":

"Despite the high information content of soil water isotope profiles, this type of data has so far rarely been included in inverse parameter identification approaches for the purpose of vadose zone modelling (Adomako et al., 2010)."

Comment:
11208, ln 12: "long time spans": there is a limit of time spans though; the expression is rather subjective and should be re-written to not cause misunderstanding. Dispersion causes mixing and therefore, the seasonal signal gets attenuated and after a certain time cannot be used to track water particles anymore.

Reply:
We substituted "long time spans" by "months to years":

"…iii) determination of pore water stable isotope concentrations allow to track water particles under variable natural boundary conditions over months to years."

Comment:
11208, ln 18, 21: "soil hydraulic properties" rather than "soil physical" (applies to entire manuscript)

Reply:
We changed this as suggested for the entire manuscript.

Comment:
11208 ln 20-23: terms like "adequate", "most reliable" are not specific; I reckon the authors want to achieve high accuracy between simulated and measured data and later see whether the identified parameters make sense for the tested soils

Reply:
We refer now to Gupta et al. (2005, Encyclopedia of Hydrological Sciences, Chapter 142) and changed the paragraph as follows:

"Does a combination of pore water isotope profiles and soil moisture time series as parallel optimization targets result in a "well calibrated" (Gupta et al., 2005) parameter

representation? Is the sequential use of soil moisture data to determine first the soil hydraulic properties and using the pore water isotope information to estimate the solute transport parameters afterwards the best way to derive a "well-calibrated" soil physical model?"

Comment:

11209, ln 21: despite the information given in Table1, more information about soil water measurements is required in the text: what sensors? how many? how close to profiles? - 11210, ln 26: how was precipitation collected?

Reply:

We included a more detailed description of the measurements of the soil data:

"The data availability varied between the study sites (Table 1). At the sites in Roodt and Eichstetten, 5TE sensors (Decagon, Pullman, USA; accuracy ±0.03 cm3 cm-3) were installed within 5 m distance to the isotope profile sampling locations for continuous soil moisture measurements that were averaged to daily values. At Roodt, the mean soil moisture content from three profiles, each with sensors at three depths (-10, -30, and -50 cm) was calculated, while no replicates were available for Eichstetten at 7 depths (-5, -10, -20, -30, -40, -50, and -60 cm). In Hartheim, the soil moisture was determined destructively with soil cores in three replicates taken weekly and in exceptions bi-weekly to three-weekly (Koeniger, 2003)."

…and precipitation data:

"Precipitation was measured either above the canopy with an ombrometer (Hartheim, Mayer et al., 2005) or in the open field with a tipping bucket (Roodt, Eichstetten). The isotopic composition of the rainfall in Roodt and Eichstetten and throughfall in Hartheim was determined at least every 14 days as bulk samples at the study sites over a period of at least 14 months before the isotope profile sampling started. At Roodt, additional event based (every 4 mm) samples were taken in 2012 and 2013, and paraffin oil was used to prevent evaporation fractionation.

Comment:

11211, ln 1: I reckon the authors analysed the water and not its vapour using the equilibrium method; be more specific here

Reply:

We changed for clarification as follows:

"The rainwater isotope analyses for Roodt and Eichstetten were done with a Wavelength-Scanned Cavity Ring Down Spectrometer (Picarro, Santa Clara, USA) that was coupled to a vaporizer to analyse liquid samples. The rain water from Hartheim was analysed with a mass spectrometer (Finnigan MAT-DeltaS, Bremen, Germany)."

Comment:
11211, ln 1-4: “minimize the influence of initial conditions” sounds awkward; to also consider the initial deuterium concentration, the time series were extended; more information is needed about these other sampling locations close by: which locations? how far were data extended? it could influence your model accuracy at the different locations

Reply:
We rephrased the sentence as suggested and included more information about the other sampling locations as follows:

“To reduce the influence of the initial conditions of the δ^2H concentration in the pore water, the time series of the isotopic composition of the precipitation were extended with additional isotope data spatially interpolated from GNIP stations as described in Seeger and Weiler (2014) for Roodt and altitude corrected from the meteorological station Schauinsland for Eichstetten.”

Comment:
11211, ln 6: Water flow instead of water transport

Reply:
We changed this as suggested.

Comment:
11212, ln 8: Why did the authors choose a LAI of 2 for the grassland sites? It seems rather low but certainly is justified if only little vegetation was present.

Reply:
Since no LAI measurements were done at the study sites, we had to apply values from the literature:

To assess the seasonal variability of the LAI in the grassland sites (Roodt and Eichstetten), the year was divided into winter season (1st of November – 1st of March, LAI = 0.2) and summer season (1st of May – 1st of September, LAI = 2) according to Breuer et al. (2003).

Comment:
11212, ln 26: I reckon the delta values + an arbitrary value were used for conversion into positive numbers because you cannot calculate with “negative” concentration data. Either this information has to be added or the sentence deleted as it could be misleading

Reply:
We changed as suggested:

The δ notation, in ‰ VSMOW of the isotopic concentration plus an offset value (to get positive values) was used for calculating the isotopic compositions and its mixing.

Comment:
11213, ln 3: add “(data not shown)”; it would be even more important if the d2H/d18O values are similar to the individual Local Meteoric Water Line

Reply:
We changed this as suggested:

This assumption was considered to have a minor impact on the simulations, because the 2H-18O relationship of the pore waters at the study sites were similar to the local meteoric water line (LMWL) below 30 cm soil depth, suggesting limited effects of isotope enrichment (data not shown).“

Comment:
11213, ln 5-9: generally, this statement is correct. If choosing the example of Maloszewski et al. (2006) it is worth mentioning that this was even for sediments without vegetation. Probably referring to one of the previously mentioned lysimeter studies containing soils and not sediments and having vegetation instead of being bare, would be even more appropriate and comparable to your sites here.

Reply:
We will refer now to the study by Stumpp et al. 2012, Vadose Zone Journal (doi: 10.2136/vzj2011.0075), who studied the percolation in lysimeter with natural soils and vegetation cover:

“Furthermore, Stumpp et al. (2012) found in a similar climate that the average deuterium contents in precipitation and the water outflow of a lysimeter in -150 cm depth were nearly the same, concluding that fractionation due to evaporation does not play a big role in temperate climates.”

Comment:
11213, ln 20: “were” instead of have been

Reply:
We changed that as suggested.

“The profiles were discretized into 101 nodes,...”

Comment:
- 11213, ln 24: “according to the soil description”: I am wondering about the choice of soil horizons at the Roodt site. it was mentioned earlier that the B horizon ends in 50 cm bgs followed by a weathered C horizon. Why not combining horizons A and B, having also similar textures, and getting a second set of flow and transport parameters for the C horizon.

Reply:
The Soil moisture time series for 30 and 50 cm depth are more similar to each other than compared to the soil moisture in 10 cm. Therefore, we decided to include the soil horizon between the A and the B horizon, rather than distinguishing between the B and C horizon. We also believe that it is crucial to get the water flow in the upper part of the soil as correct as possible, in order to keep the errors in the actual soil evaporation and root water uptake (root zone upper 20 cm) small. In addition, there was no soil moisture data for the C horizon, which would bring problems in the parameterization of the soil hydraulic parameters, because they would then depend solely on the

isotope profile data. In order to keep the number of parameters for the three study sites the same, we do not want to include a third horizon. However, we see the limitations and will consider the third horizon in a follow up study on the Cambisol in Roodt.

Comment:
11214, ln 1: a spin-up period of two years is probably not enough for the Eichstetten site when looking at the transit times (Figure 6). Here, transit times are larger than two years which needs to be discussed. It is apparently also not enough for Hartheimregarding the IPA and 2SA calculations because the deuterium content below a certain depth equals the initial average (Figure 2). Therefore, the yielded dispersivities should be taken with care and do not reflect actual dispersivities! Please add these points to the discussion

Reply:
The spin-up period has been always at least 967 days, which is longer

We included therefore the minimum number of days in section 2.2.3 and list the number of days for every site in Table 1:

"The influence of the initial conditions on the calibration can be neglected, as a spin-up period of at least 967 days was simulated before the start of the calibration period (Table 1).

and pick up this point in the discussion in section 4.2 as follows:

"Since the efficiency of the pore water isotope simulations is beside the MVG and dispersivity parameter highly dependent on the isotopic signal of the rainwater, a sufficiently long input time series is crucial in order to ensure that the initial pore water has been renewed over the simulation period to minimize the influence of the initial conditions. In our case, this is given since the spin-up periods (Table 1) are generally longer than the estimated transit times (Fig. 6)."

Comment:
11214, ln 14, Table 2: how were these initial parameter chosen? Were the initial parameters gained from PTFs in these ranges too?

Reply:
We added the following for clarification:

"The ranges of the parameter space were based on expert knowledge and are listed in Table 2."

Comment:
11215, ln 11: "(PTF)" has to be introduced here and not in ln 2, 11216

Reply:
We changed that as suggested.

Comment:
11215, ln 15-17: was an initial range for dispersivities chosen too?

Reply:
We clarified that the range of the dispersivity was not constrained as follows:

The range of the MVG parameter values of the neighbouring textural classes defined the parameter range in which the IPA was allowed to search for an optimal parameter set, while the range of the dispersivity parameter was not constrained.

Comment:
11216, ln1, 5-6: if the dispersivity was optimized, this procedure can be considered as an inverse procedure too. I was wondering why the authors have used the forward simulation procedure at all; two more sentences could be added here (e.g. PTFs most simple approach if only texture data are available); additionally, the results of the model performance are only briefly mentioned in the text later. Instead of preparing additional figures, it would still be worth giving the model efficiencies in Table 3.

Reply:
We did not consider the approach, were the dispersivity was inversely determined, but the MVG parameter were taken from the PTF, but emphasized the results of the PTF approach.

"In addition to the inverse model approaches, the efficiency of the simulations with parameter sets derived from PTFs based on soil textural information of the horizons were also tested to clarify the value of the pore water isotope data. The Rosetta PTF (Schaap et al., 2001) was used to estimate the MVG parameters and a PTF by Perfect et al. (2002) was applied for the dispersivity parameter."

Comment:
11216, ln 23-28: this functional evaluation procedure is really good to see whether differences in inversely determined parameter really lead to differences in flow and transport; 2-3 sentences on its relevance should be added here to make it easier for the reader to follow why water balance and transit time were calculated. It could also be added to the objectives of the entire study

Reply:
We included the following sentence in the objectives section to point out the benefit of the functional evaluation:

"In addition the model realism concerning water balance and transit time estimations are compared to see how much the results of the different approaches vary with regard to simulating the hydrological function of the studied soil."

Comment:
11216, ln 28: why intermediate and how were these two events chosen? arbitrarily? please be more specific

Reply:
We changed the paragraph as follows to specify why we have chosen the different events:

"To infer transit times through the soil profiles rain input was traced virtually at each study site for two events of intermediate intensities (between 8 and 13 mm day-1), one that had occurred at the beginning of October (called "fall event") and one at the beginning of May (called "spring event"). We chose intermediate rain events, because such events are big enough to generate recharge and are more representative than heavier rain events, which are less likely to occur. The two different timings were considered to cover the differences of the processes over time."

Comment:
11217, ln 22: this information (soil moisture sensors) needs to be given in the methods section

Reply:
We added the information about the uncertainty ranges in the methods section.

"At the sites in Roodt and Eichstetten, 5TE sensors (Decagon, Pullman, USA; accuracy ±0.03 $cm^3\ cm^{-3}$) were installed within 5 m distance to the isotope profile sampling locations for continuous soil moisture measurements that were averaged to daily values."

Comment:
11220, ln 6-7: Not the high Ks but rather the very low saturated water content causes the high seepage water fluxes in first place. The low saturated water content results in low effective water contents (resulting also in the short transit times, Figure 6). Hence, water more quickly reaches deeper soil regions and is evaporated or taken up by plants anymore.

Reply:
We will included the role of the low saturated water content value of the uIPA for the water balance:

"These high recharge rates, which are twice as high as the ET for Eichstetten, are due to the low saturated water content and high hydraulic conductivities in the upper soil horizon estimated by the uIPA."

and the transit time

"Pronounced differences between the approaches were found for Eichstetten, where the uIPA resulted due to the low θ_s in transit times that were two times shorter as the IPA, MOA and the 2SA (Table 5)."

Comment:
11220, ln 24-25: see previous comment

Reply:
See previous reply.

Comment:
11221, ln 9-10: it is difficult to judge the results in terms of actual processes at the study sites as no independent measurements were done; therefore, the interpretation should be limited to the comparison between modelling approaches.

Reply:
We included a comparison of the different approaches as described above and relate to this comparison as follows:

"The mean transit times (MTT) simulated with 100 randomly chosen parameter combinations from Sbest, are statistically significant different among the inverse model approaches for Eichstetten. For Roodt, transit times of the IPA and uIPA were about twice as long as for the MOA and 2SA and the latter two approaches did not differ significantly in terms of MTT. For Hartheim, the uIPA and the MOA did not differ significantly with regard to the MTT, while the others did."

Comment:
11221 ln 26-28 (and following section): it has been shown that the accuracy of ROSETTA is limited if textural classes are given only (Vereecken et al. 2010) which needs some discussion. Furthermore, I don't understand your statement why an accurateapplication of PTFs requires homogeneous flow. As you correctly mention later in the manuscript, the Richards equation assumes homogeneous flow in soils and there- fore, it is no explanation if a PTF works in one place and not in the other place.

Reply:
We rephrased the paragraph for clarification as follows:

"The inadequate representation of the soil moisture dynamics using the hydraulic properties derived with the Rosetta PTF (Table 3) shows that site-specific hydrological characteristics can hardly be reflected via textural information alone. This limited accuracy of PTFs which use only soil texture was also found in other studies as reviewed by Vereecken et al. (2010), indicating that soil structure has to be taken into account. This is especially true for Roodt, where a high rock content influences the water flow. Therefore, the application of the PTF results in a better simulation for Eichstetten and Hartheim than for Roodt, which indicates that the flow in the first two study sites is more homogenous. At Roodt, the PTF fails to represent the water flow (KGE_θ = -0.17), but the MOA and 2SA result in satisfactory simulations, showing that the inverse estimated parameters are effective parameters that hold information of non-heterogeneous flow that cannot be represented in the model.

Comment:
11222, ln 7-8: be more specific; in all approaches inverse modelling was used, but additionally having information about soil water content improved the modelling efficiency which is actually expected.

Reply:
We changed the paragraph as follows to be mmore specific

"In general the KGE_{tot} was lower in the approaches that made use of PTF than for the MOA and the 2SA, which shows the advantage of including both, the hydrometric and hydrochemical data in inverse modelling for effectively and site specifically optimizing the model parameters."

Comment:
11222 ln 14: "reasonably well"; are there any data available for comparison and to be more specific here?

Reply:
We added the following for clarification:

"Even though the soil is not a homogenous porous medium as assumed for the applied Richards equation, our simulations of water flow and isotope transport on daily resolution over several years seems to capture the hydrological processes of percolation, ET and dispersion of pore waters reasonably well in terms of soil moisture dynamics and isotope composition of the pore waters."

Comment:
11222 and 11223: see earlier comment about dispersivites at Hartheim and Eichstetten

Reply:
See the responses and clarifications regarding the spin-up periods.

Comment:
11223, ln 2-3: I would not say it is at the lower end compared to the dispersivities found in the lysimeter studies. Here, dispersivities in the soil layers were 3.9-4.7 cm (Stumpp et al. 2012) and 6.8-8.1 cm (Stumpp et al. 2009a) which is in the same order of magnitude to your results.

Reply:
We agree and changed accordingly:

The estimated values for the dispersivity parameters are mostly within the range (0.8 – 20 cm) as reported in a review by Vanderborght and Vereecken (2007) for the field scale and lysimeter studies by Stumpp et al. (2009a2012).

Comment:
11223, ln 6-7: certainly, isotope depth profiles are beneficial. However, it also requires isotope data in precipitation over long time spans which has to be available; this points should be added to the discussion and later considered in the conclusions too

Reply:
We added in the discussion in section 4.2 the following to pronounce the importance of the precipitation input data:

Since the efficiency of the pore water isotope simulations is beside the MVG and dispersivity parameter highly dependent on the isotopic signal of the rainwater, a sufficiently long input time series is crucial in order to ensure that the initial pore water has been renewed over the simulation period to minimize the influence of the initial conditions. In our case, this is given since the spin-up periods (Table 1) are generally longer than the estimated transit times (Fig. 6).

Comment:
11223, ln 8-19: It was difficult reading this section and following your thoughts.
Reply:
We rephrased and erased parts as follows for clarification:

"In addition, only 1 to 2 sampling campaigns are necessary to get the additional information for water and solute transport. The high variability of the dispersivity between the sites and horizons in our study and reported in other studies (Vanderborght and Vereecken, 2007) and the limited model efficiencies when PTFs were applied emphasize the importance to consider the dispersivity in the parameterization of soil physical models. A field scale representation of the dispersion processes cannot be assumed for a certain soil texture by a PTF, but should rather be derived for the particular field site. "

Comment:
11225, section of transit times: I was missing some discussion on the water balance calculations here too. Does the functional evaluation now show that results are all similar anyway - no matter of soil hydraulic properties when also considering the uncertainties? Or are there crucial differences in flow and transport?

Reply:
We added in the results section the statistics on the similarity between the approaches regarding the water balance and transit times. Additionally we discuss in section 4.4 these results as follows:

"The simulated transit time distributions reveal that the water transport can differ by several weeks to months, depending on the inverse modelling approach, while the water balance estimations seem to be less sensitive to the method used to derived the parameter sets (except for the uIPA). Besides the timing of the tracer breakthrough, also the amount of recharge is sensitive to the estimated parameter set as shown in the deviation between maximum actual cumulative recharge and total possible recharge (= 1 in the cumulative density functions in

Fig. 6). Thus, our study showed that the parameter estimation for soil physical models is more crucial for transit time modelling than for water balance calculations."

Comment:
Table 1: soil moisture data, Hartheim: check the spelling of "-30"; the second horizon in Roodt ends in 50cm and therefore it is not >25 cm; do you have any information about C(v) horizons at the other sites?

Reply:
The spelling was corrected. As already stated above, the first horizon at Roodt, the A-horizon, ends in 25 cm depth and so does the first layer in the set up of the model. On the decision how to subdivide the soil profile into layers, we refer to the reply above.

Comment:
Table 1: please add the maximum root depth in the table

Reply:
We added the maximum rooting depth.

Comment:
Figure 1: please change the range of the y-axis for Hartheim (e.g. 0-0.6) to better see differences/similarities between observed and simulated values; additionally, it is difficult to see some of the overlapping simulated soil moisture curves (e.g. 30 and 50 cm in Roodt, uIPA)

Reply:
We adjusted the y-axis accordingly and changed the curves (by dashed lines) to make the differentiation between the different simulated soil moisture time series easier.

Comment:
Figure 2: uIPA Hartheim: what is the reason for the oscillations in the deeper part of the profile? Is it possible that these are numerical oscillations?

Reply:
The reason of the oscillation was a numerical oscillation, which was solved with a smaller space between the nodes.

Comment:
Figure 3: please indicate in the title what white and dark green means; the darker the more narrow is the parameter range - did I get it right?

Reply:
We changed the in accordance to the new sensitivity analysis and added to the caption the following:

"Green indicates a small range, yellow medium and orange represents a high range."

Comment:
Figure 5 and 6: please indicate in the title that it refers to seepage water fluxes and transit times in 200cm depth

Reply:

We will indicated in the caption that the recharge fluxes and transit time calculations refer to 200 cm depth for Roodt and Eichstetten and to 120 cm for Hartheim.

Author's response to the review by Referee #2

We thank the Referee #2 for the thoughtful comments and provide the changes that were done and/or detailed replies in the following responses.

Comment:
First, it should be made clearer what the advantages of using profiles of isotope concentrations instead of profiles of an inert tracer substance are. Especially since the measurement of isotope concentrations and the determination of the boundary conditions are much more complicated, it is important to point out the advantages of this method. In this respect, it could be useful to refer to novel experimental procedures that allow to determine these profiles online in the field ([Rothfuss et al., 2013]. Such an online method allows obtaining profiles with much higher temporal resolution, which might also provide important additional information that allows constraining soil parameters better.

Reply:
We included the following paragraph in section 1.2 to show with regard to reviewed literature the advantages of the applied method:

"These and other studies have shown the advantages of stable water isotopes over inert tracers either naturally or artificially introduced. One major benefit is that several hydrological processes which take place over longer time spans, such as infiltration, evaporation, transpiration, percolation, are integrated in the shape of the pore water stable isotope profiles. Thus, pore water stable isotope data provides information of natural processes that occur during different hydrological states (e.g. wet or dry periods). Especially, the fact that stable isotopes are part of the water molecule and therefore extracted (without fractionation) via root water uptake is helpful to constrain transpiration, which would not be possible with an artificial tracer. Recently developed laboratory methods allow to determine the stable isotope composition of soil samples time efficient at high precision (Wassenaar et al., 2008) and novel in-situ measurements make the sampling of pore water stable isotopes even more convenient (Rothfuss et al., 2013; Volkmann and Weiler, 2014). Last but not least, pore water stable isotopes provide the means to include the transport parameter (diffusivity) into inverse modelling approaches, which would not be possible with solely water content or matric potential data."

Comment:
Second, the text is at several points unclear and the methods are not sufficiently well explained. Crucial information about the measurement setup is missing in the results section it would be good to include information about the pedotransfer functions and the obtained parameters.

Reply:
We have included a more detailed description of the methods with special emphasis on the measurement setup:

"The data availability varied between the study sites (Table 1). At the sites in Roodt and Eichstetten, 5TE sensors (Decagon, Pullman, USA; accuracy ±0.03 cm^3 cm^{-3}) were installed within 5 m distance to the isotope profile sampling locations for continuous soil moisture

measurements that were averaged to daily values. At Roodt, the mean soil moisture content from three profiles, each with sensors at three depths (-10, -30, and -50 cm) was calculated, while no replicates were available for Eichstetten at 7 depths (-5, -10, -20, -30, -40, -50, and -60 cm). In Hartheim, the soil moisture was determined destructively with soil cores in three replicates taken weekly and in exceptions bi-weekly to three-weekly (Koeniger, 2003)."

We included the results of the PTF in the results and discussion chapters and also in the Table 3 and 4.

Comment:
Third, the authors argue that they determine parameters of the soil system that are relevant for a larger scale than the scale of soil columns that are investigated in the lab. However, I disagree with this statement since the data they use are still point data which do not have a larger support volume than the scale of lab column. This problem could be circumvented if information from many point measurements at a large number of locations is combined.

Reply:
The field measurements hold information about the natural processes that happen in the field. Such conditions, as naturally developed soil structures, vegetation influences, natural time variable boundary conditions cannot or only hardly be replicated in the laboratory with soil columns. We believe to better capture these kind of processes with our field measurements of soil moisture and pore water stable isotope data. However, we agree that the soil cores are only capturing point data, but we believe that it is relative easy to measure at several locations to capture the small and larger scale variability.

Comment:
Finally, I did not understand the sensitivity analysis that was carried out and I think that an uncertainty analysis of the obtained hydraulic properties and predicted seepage, annual evapotranspiration rates is necessary.

Reply:
We have changed the sensitivity analysis as follows:

"As a sensitivity analysis, the set of model runs of the optimization process were considered whose deviation from the best run in terms of KGE was not more than 0.05 (S_{best} with $KGE_i > (KGE_{best} - 0.05)$). Of this selection the 10 to 90 percentile range (PR_{10-90}) was calculated."

We further applied this sensitivity analysis for the consequences of the water balacnce and transit time estimations:

"The sensitivities of the different approaches with regard to the water balance and transit time estimations were tested with simulations of 100 randomly chosen parameter sets from Sbest. " The results are visualized in the Figures 3,5, and 6.

Comment:
P 11205 ln12: What is meant by "transforming" water and solutes. How can water and substances be 'transformed'?

Reply:
As indicated, we refer to Blum (2005), where the functions of soils are discussed. The original sentence is: “Filtering, buffering and transformation capacity between the atmosphere, the ground water and the plant cover, strongly influencing the water cycle at the earth surface, as well as the gas exchange between terrestrial and atmospheric systems and protecting the environment including human beings, against the contamination of ground water and the food chain.” (Blum, 2005, section 1.1.2). Transforming is meant in a way where water is being transferred/converted between the different environmental compartments. Within these transformations, soil plays a major role.

Comment:
P 11207: ln1-2: I think that the authors misinterpreted the results of Vanderborght and Vereecken here. In figures 4 and 5 of Vanderborght and Vereecken, there is not a difference between dispersivities derived from column or field scale experiments. The important factor seems to be the transport distance. As long as the soil columns are long enough, parameters that are relevant for field conditions could also be obtained from column scale experiment.

Reply:
It is true that there are no differences within the same range of the travel distances, but between the travel distances (see, Fig. 6 in Vanderborght and Vereecken, 2007). Thus, it is crucial to derive the diserpsivity parameter on the scale of interest, which would not be the case for soil cores. Therefore, we will change the sentence in a way to focus on the differences between soil cores and field data. However, Vanderborght and Vereecken (2007) state that “lateral redistribution may take place cross a larger distance so that field-scale dispersion was larger than the dispersion observed in column-scale studies. Therefore, column-scale studies in coarse-textured soils,[…], may not be representative for field-scale dispersion,[…]” (Vanderborght and Vereecken, 2007). We changed this part as follows:

“For the transport parameters, experiments at the field scale are expected to be more representative of the real conditions than studies at soil cores, because of the scale dependency of the longitudinal dispersivity (Vanderborght and Vereecken, 2007). ”

Comment:
P 11208: ln 3-5: ‘Despite the high information content of soil water isotope profiles, this type of data has so far not been included in inverse parameter identification approaches for the purpose of vadose zone modelling.’ I would like to bring to the authors’ attention two papers by Mathieu Javaux who analyzed chloride tracer profiles in a deep vadose zone to derive vadose zone transport parameters[Javaux and Vanclooster, 2004a; b]. The problem that was dealt with in these papers is similar to the analysis of water isotope profiles since also non-controlled variations of chloride in the input water were used to interpret time series of concentration measurements at different depths.

Reply:
The studies by Javaux and Vanclooster (2004a; b) used an extremely labor and cost intensive experimental set up, which has only little to do with the pore water extraction method that we present. We present in our study that a much easier experimental set up results in valuable information for inverse modeling approaches that include the transport parameters and not only the soil hydraulic parameters. We show that taking two to several isotope profiles, instead of generating a time series like Javaux and Vanclooster (2004a; b), is helpful to derive water flow and transport parameters. In contrast to Javaux and Vanclooster (2004a; b), we did not use porous cups to extract

the water, but the direct equilibration method by Wassenaar et al. (2008), which provides tracer information of a wider pore spectrum than suction lysimeter samples with a constant suction of 80 kPa. Because of the listed differences, we don't see a lot of parallels to our study and did not include the studies in our manuscript.

Comment:
P 11208 ln 9-10: '(ii) parameter optimization/estimation should be conducted on the scale of the application.' I agree with this statement but the critical question is whether the observations represent the scale of application. If isotope concentration profiles are determined at the local scale, i.e. a small volume around a suction cup, then it is questionable whether these measurements are representative for a larger scale. The same holds true for soil water content measurements. If water contents are measured only at a single location with a sensor that has a small sampling volume (such as the 5TE sensors) then it is also questionable whether this measurement is relevant for a larger scale.

Reply:
Obviously, there is the issue of the heterogeneity of the soil all soil physicists have to deal with. To account for the heterogeneity, we averaged three soil moisture sensors at each depth for the study site in Roodt. For the other sites, there were no replicate measurements available. For the pore water stable isotope profiles, there were usually only one profile taken. However, we included as an example of the variabilty a study conducted at the site in Eichstetten:

"No replicates of the isotope profiles were available in this study, but it was shown at Eichstetten that the interquartile range was smaller than 1.5 ‰ for the pore water δ^2H at the same depths for 10 isotope profiles taken in parallel (Eisele, 2013), which is similar to the measurement accuracy."

Comment:
P 11209: 'slightly clayey silt' and 'silty sandy': use correct nomenclature for soil texture classes. These texture classes do not exist in the USDA textural triangle.

Reply:
We changed soil texture classes according to the USDA textural triangle into "silt" and "silt loam".

Comment:
P 11209: ln 18-19 'All three sites are located on undulating terrain, where vertical flow is dominating and lateral subsurface flows can be neglected.' Give the maximal slopes. Given that the soils are relatively shallow in the Roodt catchment, I am wondering whether the weathered schist does not lead to perched water tables and lateral subsurface flow.

Reply:
We will included the information that the slope is at all the sites below 3°:

"All three sites are located on undulating terrain (slopes <3°), where vertical flow is dominating and lateral subsurface flows can be neglected."

Comment:
P 11209 ln 20- p 11210 ln 5: Which soil sensors were installed, at which depths, how many

repetitions per depth how far were the soil sensors separated from each other? How many soil samples were taken to determine the water content profiles, how many profiles were taken for the isotope concentration measurements, what was the size of the cores, how far where the cores from the location of the soil moisture sensors? Part of this information is in table 1 but not everything. Table 1 should be referred to in the text.

Reply:
We have changed that part as follows:

"The data availability varied between the study sites (Table 1). At the sites in Roodt and Eichstetten, 5TE sensors (Decagon, Pullman, USA; accuracy ±0.03 cm^3 cm^{-3}) were installed within 5 m distance to the isotope profile sampling locations for continuous soil moisture measurements that were averaged to daily values. At Roodt, the mean soil moisture content from three profiles, each with sensors at three depths (-10, -30, and -50 cm) was calculated, while no replicates were available for Eichstetten at 7 depths (-5, -10, -20, -30, -40, -50, and -60 cm). In Hartheim, the soil moisture was determined destructively with soil cores in three replicates taken weekly and in exceptions bi-weekly to three-weekly (Koeniger, 2003). Also the Table 1 was changed accordingly.

Comment:
P 11210: ln 27-29: 'The isotopic composition of the rainfall in Roodt and Eichstetten and throughfall in Hartheim was determined at the study sites at least 14 months before the isotope profile sampling started and then at least every 14 days' I did not understand this. Do you mean that the at least 14 months before the isotope sampling, isotope composition of rain/throughfall water was determined at least every 14 days.

Reply:
We changed this section for clarification as follows:

"Precipitation was measured either above the canopy with an ombrometer (Hartheim, Mayer et al., 2005) or in the open field with a tipping bucket (Roodt, Eichstetten). The isotopic composition of the rainfall in Roodt and Eichstetten and throughfall in Hartheim was determined at least every 14 days as bulk samples at the study sites over a period of at least 14 months before the isotope profile sampling started. At Roodt, additional event based (every 4 mm) samples were taken in 2012 and 2013, and paraffin oil was used to prevent evaporation fractionation."

Comment:
P 11211: ln 1-3: 'To minimize the influence of the initial conditions of the deuterium concentration in the pore water, the time series of isotope concentration of the precipitation were extended with additional isotope data from other sampling locations close by.' I did not understand this: in what sense was the isotope concentration of precipitation 'extended'?

Reply:
We changed this part for clarification as follows:

"To reduce the influence of the initial conditions of the δ^2H concentration in the pore water, the time series of the isotopic composition of the precipitation were extended with additional

isotope data spatially interpolated from GNIP stations as described in Seeger and Weiler (2014) for Roodt and altitude corrected from the meteorological station Schauinsland for Eichstetten.“

Comment:
P 11214 ln 2-5: The definition of the upper boundary conditions is not precise enough. First, the upper boundary condition at the soil profile is not governed by the evapotranspiration since the evapotranspiration includes both evaporation from the soil surface and transpiration from the canopy. Second, it is not clear how the boundary condition for the Deuterium is set when evaporation occurs. I suppose that a zero concentration of Deuterium at the soil surface is set when evaporation occurs and a third type boundary condition when infiltration with a known concentration occurs.

Reply:
We changed this part as follows to clarify the upper boundary condition:

“The upper boundary condition was defined by variable atmospheric conditions (Cauchy boundary condition) that govern the loss of water and deuterium caused by evaporation, the input of water due to throughfall and the accompanied flux concentrations of deuterium. Since we use a modified version of the HYDRUS code (Stumpp et al. 2012), evaporation influences only the amount of water, not its isotopic composition. The lower boundary was set to zero-gradient with free drainage of water and solutes.“

Comment:
P 11216: The parameter space was not unconstrained in the other cases. It was constrained by preset ranges that were derived based on expert knowledge.

Reply:
We changed this part as follows:

“The range of the MVG parameter values of the neighbouring textural classes defined the parameter range in which the IPA was allowed to search for an optimal parameter set, while the range of the dispersivity parameter was not constrained. Also an alternative, where the parameter space of the MVG was not constrained based on expert knowledge (unconstrained) was tested (uIPA).“

Comment:
P 11216: I don’t think that the sensitivity analysis that is presented is appropriate. Since the SCE-UA algorithm looks for the best parameter set, the distribution of the parameter sets that are obtained do not represent a posterior parameter distribution. The question is whether the distribution will become ergodic or reach a steady variance if always more and more parameter sets are considered. If this is not the case but if the distribution always becomes narrower and narrower around the optimum parameter set when more and more parameter sets are evaluated in the monte-carlo chain, then the width of the distribution depends of the number of parameter sets that were considered in the monte carlo chain. The width of the distribution of the 10% best parameter sets will then depend also on the number of parameter sets that were evaluated in the Monte-Carlo chain and not only on the sensitivity of the parameter.

Reply:
We changed the sensitivity analysis as follows:

"As a sensitivity analysis, the set of model runs of the optimization process were considered whose deviation from the best run in terms of KGE was not more than 0.05 (S_{best} with $KGE_i > (KGE_{best} - 0.05)$). Of this selection the 10 to 90 percentile range (PR_{10-90}) was calculated."

and extended the sensitivity analysis as follows:

"The sensitivities of the different approaches with regard to the water balance and transit time estimations were tested with simulations of 100 randomly chosen parameter sets from Sbest. If the different inversely determined parameter sets lead to significant different functional responses with regard to flow and transport was tested with a one-way ANOVA and a Post-hoc analysis (Tukey's HSD). The tested variables were the mean annual ET and the median transit time, defined as the time after which half of the recharge water has passed the lower boundary of the soil profile. "

Comment:
P 11217 ln 1: If only two events are considered, why are the rain intensities of the events 'between' 8 and 13mm d-1 considered then? I would say that the rain intensities were 8 or 13 mm d-1. Or were several applications in different years in the beginning of October or the beginning of May considered?

Reply:
We clarified this part as follows:

"To infer transit times through the soil profiles rain input was traced virtually at each study site for two events of intermediate intensities (between 8 and 13 mm day^{-1}), one that had occurred at the beginning of October (called "fall event") and one at the beginning of May (called "spring event"). We chose intermediate rain events, because such events are big enough to generate recharge and are more representative than heavier rain events, which are less likely to occur. The two different timings were considered to cover the differences of the processes over time."

Comment:
Chapter 3.1: Simulation results using parameters derived directly from pedotransfer functions are discussed. But, I would propose to include the parameters derived from pedotransfer functions also in a table and maybe also show the hydraulic functions that were obtained from pedotransfer functions in figure 4.

Reply:
Since the simulations with the PTF do not result in a comparable good agreement as the other approaches do, we passed on a visualization of these data. We will still do so in a revised manuscript, because the additional information is limited on the cost of lucidity. However, we have included the results of the PTF in Table 3 and also listed the parameter in Table 4.

Estimating flow and transport parameters in the unsaturated zone with pore water stable isotopes

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Abstract

Determining the soil hydraulic properties is a prerequisite to physically model transient water flow and solute transport in the vadose zone. Estimating these properties by inverse modelling techniques has become more common within the last two decades. While these inverse approaches usually fit simulations to hydrometric data, we expanded the methodology by using independent information about the stable isotope composition of the soil pore water depth profile as a single or additional optimization target. To demonstrate the potential and limits of this approach, we compared the results of three inverse modelling strategies where the fitting targets were a) pore water isotope concentrations, b) a combination of pore water isotope concentrations and soil moisture time series, and c) a two-step approach using first soil moisture data to determine water flow parameters and then the pore water stable isotope concentrations to estimate the solute transport parameters. The analyses were conducted at three study sites with different soil properties and vegetation. The transient unsaturated water flow was simulated by ~~numerically~~ solving the Richards equation numerically with the finite-element code of HYDRUS~~Hydrus~~-1D. The transport of deuterium was simulated with the advection-dispersion equation, and a modified version of HYDRUS was used, allowing deuterium loss during evaporation~~the Hydrus code was modified to allow for deuterium loss during evaporation~~. The Mualem-van Genuchten and the longitudinal dispersivity parameters were determined for two major soil horizons at each site. The results show that approach a) using only the pore water isotope content cannot substitute hydrometric information to derive parameter sets that reflect the observed soil moisture dynamics, but gives comparable results when the parameter space is constrained by pedotransfer functions. Approaches b) and c) using both, the isotope profiles and the soil moisture time series resulted in ~~satisfying model performances~~good simulation results with regard to the Kling-Gupta-Efficiency and good

parameter identifiability. However, approach b) has the advantage that it considers the isotope data not only for the solute transport parameters, but also for water flow and root water uptake, and thus increases parameter realism. Approaches b) and c) both outcompeted simulations run with parameters derived from pedotransfer functions, which did not result in ~~satisfying model efficiencies~~an acceptable representation of the soil moisture dynamics and pore water stable isotope composition. Overall, parameters based on this new approach that includes isotope data lead to similar model performances regarding the water balance and soil moisture dynamics and better parameter identifiability than the conventional inverse model approaches limited to hydrometric fitting targets. If only data from isotope profiles in combination with textural information is available, the results are still satisfactory. This method has the additional advantage that it will not only allow us to estimate water balance and response times, but also site-~~ ~~specific time variant transit times or solute breakthrough within the soil profile.

1. Introduction

1.1. Inverse modelling

Soils play a major role in the water cycle due to their capacity for filtering, buffering and transforming water and solutes between the atmosphere, the ground water and the vegetation cover (Blum, 2005). Soil physical models are widely used to describe water flow and solute transport in the vadose zone, for example to estimate groundwater recharge and the resulting leaching of solutes (e.g. Vanclooster et al., 2004; Christiansen et al., 2006) and the effects of climate variability (Strasser and Mauser, 2001) and climatic extremes (Bormann, 2009, 2012) on the soil water balance. However, determining the crucial model parameters describing the soil hydraulic functions (Gribb et al., 2009) and solute transport remains a challenge because of the pronounced spatial heterogeneity (Corwin et al., 2006). Methods to determine soil hydraulic characteristics include laboratory measurements of the water retention curve or the hydraulic conductivity of a particular soil sample or soil core or pedo-transfer functions based on grain size distributions (Vereecken et al., 2010). Moving beyond the point scale, the inverse model approach allows optimizing the model parameters by fitting model simulations to observed data at the scale of interest (Russo et al., 1991; Durner et al., 1999; Hopmans et al., 2002; Vrugt et al., 2008). These scales range from soil column experiments in the lab where water content, matric potentials and outflow were measured and then used for the

parameterization of numerical models (e.g. Whisler and Watson 1968) to the field scale (e.g. Dane and Hruska 1983).

Extending the inverse modelling approach by using a combination of different types of data as objective functions generally improves parameter identification (Kool et al., 1985, Ritter et al., 2003). For example, a combination of hydrometric and hydro-chemical data allows to optimize both the parameters governing water flow and solute transport, while reducing the ill-posedness of inverse problems (Mishra and Parker, 1989; Medina et al., 1990; Russo et al., 1991). Since transient unsaturated flow and solute transport processes are coupled, two possible approaches to the inverse problem were identified: a simultaneous or a sequential approach, in which hydrometric (e.g. soil moisture, matric potential, outflow) and tracer data (e.g. concentrations in the outflow) are used to either determine the soil hydraulic parameters and the transport parameters in parallel or in two steps (Mishra and Parker, 1989). Mishra and Parker (1989) found that the simultaneous optimization yielded lower parameter uncertainties than the sequential method. The simultaneous optimization approach was applied to infer water flow and solute transport parameters from tracer experiments in columns (Inoue et al., 2000) and at the field scale (Jacques et al., 2002; Abbasi et al., 2003a, 2003b). The sequential approach was used in lysimeter studies under natural conditions, with cumulative outflow and its stable isotope concentration serving as objective functions for the water flow (Maciejewski et al., 2006) and transport parameters (Maloszewski et al., 2006).

While soil core/column and lysimeter experiments have the advantage of well-known boundary conditions, their suitability to derive soil properties for predicting field-scale processes is questionable (Russo et al., 1991). Comparative studies showed that the soil hydraulic properties~~physical parameters~~ derived from inverse modelling on the scale of the targeted model application outcompete parameter sets resulting from laboratory experiments (Ritter et al., 2003; Kumar et al., 2010; Kuntz et al., 2011). For the transport parameters, experiments at the field scale are expected to be more representative of the real conditions than studies at soil ~~column or lysimeter studies~~cores, because of the scale dependency of the longitudinal dispersivity (Vanderborght and Vereecken, 2007). The inverse modelling approach on the field scale generally results in effective parameters, which lump the systems subscale heterogeneity and describe its behaviour at the targeted scale (Pachepsky et al., 2004).

1.2. Pore water stable isotope profiles

As mentioned above, including hydro-chemical data into the inverse modelling approach has distinct advantages. The concentration of stable water isotopes in the stream flow have widely been used to improve calibration and realism of catchment models (e.g. Birkel et al., 2011; Hartmann et al., 2012) and to infer transit times or residence times of catchments (e.g. Maloszewski et al., 1983; McGuire and McDonnell, 2006; Maloszewski et al., 1992; Fenicia et al., 2010; Roa-Garcia and Weiler, 2010; Birkel et al., 2012; Seeger and Weiler, 2014). Similarly, the concentration of stable isotopes in the outflow of lysimeters where used to derive transit times in the vadose zone (Stumpp et al., 2009a, 2009b). However, this type of flow concentration data is not easy to come by at the pedon scale, where we usually are not able to measure breakthrough curves, as we would do in column or lysimeter experiments. One possible solution to this problem is the determination of stable water isotopes (deuterium (^{2}H) and oxygen-18 (^{18}O)) in the pore water. If the isotopic composition of the infiltrating water varies over time, the water transport within a soil profile can thus be traced. Hence, the time dimension of the tracer input (isotopes in the rain over a several year sequence) is preserved in the space dimension (isotopes in the pore water over depth) (Eichler, 1966).

Such pore water stable isotope analyses have shown to give valuable insights into the hydrological processes in the vadose zone of temperate regions~~: these profiles~~ providinge information: on the water balance of forest soils (Eichler, 1966; Zimmermann et al., 1966; Blume et al., 1967; Wellings, 1984) and the infiltration and percolation processes (Darling and Bath, 1988; Gazis and Feng, 2004; Koeniger et al., 2010; Thomas et al., 2013), on the influence of vegetation on evaporation (Zimmermann et al., 1967), on preferential root water uptake (Gehrels et al., 1998), and on subsurface hydrological processes in hillslopes (Blume et al., 1968; Garvelmann et al., 2012). These and other studies have shown the advantages of stable water isotopes over inert tracers either naturally or artificially introduced. One major benefit is that several hydrological processes which take place over longer time spans, ~~like~~such as infiltration, evaporation, transpiration, percolation, are integrated in the shape of the pore water stable isotope profiles. Thus, pore water stable isotope data provides information of natural processes that occur during different hydrological states (e.g. wet or dry periods). Especially, the fact that stable isotopes are part of the water molecule and therefore extracted (without fractionation) via root water uptake is helpful to constrain transpiration, which would not be possible with an artificial tracer. Recently developed laboratory methods allow to determine the stable isotope composition of soil samples time efficient at high precision (Wassenaar et al., 2008) and novel in-situ measurements make the

sampling of pore water stable isotopes even more convenient (Rothfuss et al., 2013; Volkmann and Weiler, 2014). Last but not least, pore water stable isotopes provide the means to include the transport parameter (diffusivity) into inverse modelling approaches, which would not be possible with solely water content or matric potential data. Despite the high information content of soil water isotope profiles, this type of data has so far ~~not~~ rarely been included in inverse parameter identification approaches for the purpose of vadose zone modelling (Adomako et al., 2010).

1.3. Objectives

Previous work can be summarized in the following statements which guided the design of our study: i) a combination of hydrometric and hydro-chemical data decreases ill-posedness of an inverse problem, ii) parameter optimization/estimation should be conducted on the scale of the application, iii) determination of pore water stable isotope concentrations allow to track water particles under variable natural boundary conditions over ~~long time spans~~months to years. As mentioned above, the use of pore water stable isotope profiles for calibration of soil ~~physical parameters~~hydraulic properties for the vadose zone in a humid climate has so far not been rigorously tested. This study will fill this research gap by focusing on three different approaches to include pore water isotope concentrations in an inverse modelling framework and thus answering the following research questions: Do stable water isotope profiles as a solitary optimization target provide enough information to derive soil hydraulic properties~~physical~~ and solute transport parameters? Does a combination of pore water isotope profiles and soil moisture time series as parallel optimization targets result in a ~~more adequate~~"well calibrated" (Gupta et al., 2005) parameter representation? Is the sequential use of soil moisture data to determine first the soil hydraulic properties~~physical parameters~~ and using the pore water isotope information to estimate the solute transport parameters afterwards - the best way to derive a "well-calibrated" soil physical model?~~most reliable?~~ The objective of this paper is to investigate these questions in a comparative study applying all optimization approaches to three different sites and thus a range of soil types. The different inverse model approaches that include either pore water stable isotope concentrations alone or in combination with soil moisture data in a parallel or subsequent manner will be compared with regard to the model performances~~,~~ and their parameter identifiability. In addition~~and~~ the model realism concerning water balance and transit time estimations are compared to see how much the results of the different approaches vary with regard to simulating the hydrological function of the studied soil.

2. Methods

2.1. Site descriptions and data availability

The inverse model approaches were tested for three study sites located in temperate central Europe: Roodt, in the West of the Grand Duchy of Luxemburg, and Eichstetten and Hartheim, in the Southwest of Germany. Their environmental characteristics and available data are summarized in Table 1. The three study sites have a similar climate, with rainfall occurring all year with mean precipitation between 660 to 900 mm yr^{-1}. However, the study sites differ in their geological and pedological setting. The soil in Roodt is a Cambisol characterized by a ploughed humous mineral horizon (Ap) in the upper 25 cm, followed by a loamy brown B-horizon (Bv) over heavily weathered schist rocks (stone content >80%; Cv) starting in 50 cm soil depth. In Eichstetten, the prevailing soil is a ~~slightly clayey~~ silt Luvisol, developed on pleistocene aeolian loess (Hädrich and Stahr, 2001). In Hartheim, the soil is a Calcaric Regosol with a silt~~y sandy~~ loam top soil (>40 cm) on fluvial gravel and coarse sand (Schäfer, 1977). The study sites in Roodt and Eichstetten are grasslands and the site in Hartheim is a Scots pine plantation (*Pinus sylvestris*). All three sites are located on undulating terrain (slopes <3°), where vertical flow is dominating and lateral subsurface flows can be neglected.

The data availability varied between the study sites (Table 1). At ~~T~~the sites in Roodt and Eichstetten, 5TE sensors (Decagon, Pullman, USA; accuracy ~~(~~±0.03 cm^3 cm^{-3}~~)~~) were installed within 5 m distance to the isotope profile sampling locations for~~had~~ continuous soil moisture measurements that were averaged to daily values. At Roodt, the ~~a~~ mean soil moisture content from three profiles, each with sensors at three depths (-10, -30, and -50 cm) was calculated, while no replicates were available for Eichstetten at 7 depths (-5, -10, -20, -30, -40, -50, and -60 cm).~~, whereas i~~In Hartheim, the soil moisture was determined destructively with soil cores in three replicates taken weekly and in exceptions bi-weekly to three-weekly (Koeniger, 2003). The methodology for the pore water isotope measurements differed for the different study sites, due to the technical possibilities at the time of the sampling. At the sites in Roodt and Eichstetten, the soil samples were taken during the years 2012 and 2013 and analysed for their pore water isotopic composition according to the equilibration method (Wassenaar et al., 2008). Each isotope profile was determined by taking soil samples in 5 cm depth intervals from a soil core of 8 cm diameter excavated with a percussion drill (Atlas Copco Cobra). The soil samples were taken to the laboratory in sealed air-tight bags. In addition to the soil samples, standards were prepared, which consisted of oven-dried soil material that was rewetted to the soil moisture at the time of sampling with three different waters of known

isotopic composition. After adding dry air to both, standards and field samples, the bags were re-sealed. The soil pore water was allowed to equilibrate with the dry atmosphere in the bag for two days under constant temperature (21°C). The headspace in the bags was directly sampled with a Wavelength-Scanned Cavity Ring Down Spectrometer (Picarro, Santa Clara, USA) for 6 minutes, and only the measured concentration of ~~deuterium~~ ^{2}H and ^{18}O during the last ~~90~~ 120 seconds was averaged to minimize carryover effects. The isotopic composition of the gas phase was converted to values of the liquid pore water according to the temperature dependent fractionation factor as defined by Majoube (1971). The standards were measured at the beginning, every three hours during, and at the end of the analysis for each profile. The standards were used to account for drift of the laser spectrometer and to calibrate the measurements in order to get values in the δ--notation relative to the Vienna Standard Mean Ocean Water (VSMOW). The measurement accuracy, given as the average range of repeated measurements of the standards over the day, was 1.~~96~~ 45 ‰ for δ ~~Deuterium~~ ^{2}H. At the Hartheim site, the sampling took place in 1999 and 2000 and the pore water isotope analysis was done by excavating 500 g of soil in 5 cm intervals and extracting the pore water with the means of azeotropic distillation with toluol (Koeniger, 2003; Revesz and Woods, 1990). The extracted pore water was then analysed for the ^{2}H~~deuterium~~ concentration with a mass spectrometer (Finnigan MAT-DeltaS, Bremen, Germany). No replicates of the isotope profiles were available in this study, but it was shown at Eichstetten that the interquartile range was smaller than 1.5 ‰ for the pore water $\delta^{2}H$ at the same depths for 10 isotope profiles taken in parallel (Eisele, 2013), which is similar to the measurement accuracy.

Precipitation was measured either above the canopy with an ombrometer (Hartheim, Mayer et al., 2005) or in the open field with a tipping bucket (Roodt, Eichstetten). The isotopic composition of the rainfall in Roodt and Eichstetten and throughfall in Hartheim was determined at least every 14 days as bulk samples at the study sites over a period of at least 14 months before the isotope profile sampling started ~~and then at least every 14 days~~. At Roodt, additional event based (every 4 mm) samples were taken in 2012 and 2013, and paraffin oil was used to prevent evaporation fractionation. The rainwater isotope analyses for Roodt and Eichstetten were done with a Wavelength-Scanned Cavity Ring Down Spectrometer (Picarro, Santa Clara, USA) that was coupled to a vaporizer to analyse liquid samples. The rain water from Hartheim was analysed with a mass spectrometer (Finnigan MAT-DeltaS, Bremen, Germany) ~~the same equipment as the pore water analysis~~. To ~~minimize~~ reduce the influence of the initial conditions of the $\delta^{2}H$~~deuterium~~ concentration in the pore water, the time series of the isotopic ~~concentration~~ composition of the precipitation were extended with additional

isotope data spatially interpolated from ~~other~~ GNIP stations as described in Seeger and Weiler (2014) for Roodt and altitude corrected from the meteorological station Schauinsland for Eichstetten~~sampling locations close by~~. Although, the isotope analysis were done for δ^2H and $\delta^{18}O$, we only consider δ^2H in the inverse modelling approaches, because i) the relative errors of the stable isotope analysis were smaller for δ^2H with a standard deviation of 1.16‰ compared to 0.31‰ for $\delta^{18}O$, ii) ^2H is less affected by fractionation processes than ^{18}O, iii) the additional gain of information of considering both isotopes vs. just ^2H is limited, since $\delta^{18}O$ and δ^2H are highly correlated, and iv) the HYDRUS model cannot account for fractionation processes due to evaporation.

2.2. Model setup

2.2.1. Water ~~transport~~flow

The transient water flow within the unsaturated soil profile was simulated by numerically solving the Richards equation with the finite-element code of ~~Hydrus~~HYDRUS-1D (Šimůnek et al., 2012). For the parameterization of the water retention ($\Theta(h)$) and the unsaturated hydraulic conductivity ($K(h)$) functions, the Mualem-van Genuchten model (van Genuchten, 1980) was applied. These relations are specified by the residual and saturated volumetric water contents (θ_r [L^3 L^{-3}] and θ_s [L^3 L^{-3}], respectively), the inverse of the capillary fringe thickness (α [L^{-1}]), two shape parameters (n [-], and m [-], where m = 1-1/n), the saturated hydraulic conductivity (K_s [L T^{-1}]), and a tortuosity parameter (l [-], in accordance to Mualem (1976) set to 0.5 to reduce the number of free parameters).

A sink term in the Richards equation was defined according to the root water uptake model by Feddes et al. (1978), which describes the reduction of the potential water uptake by a dimensionless trapezoidal stress response function. Such non-optimal conditions for the vegetation are defined by pressure heads above and below which the plants experience oxygen or water stress, respectively. In this study, the following prescribed parameter set for pasture (Wesseling, 1991) was used for all sites, since no parameter for scots pine are available: >-10 cm oxygen stress occurs; between -25 and -800 cm optimum; below -8000 cm root water uptake ceases. The root water uptake was restricted to the root zone, which was defined by the sites' specific rooting depth (20 cm, 30 cm, and 40 cm for Roodt, Eichstetten, and Hartheim, respectively) and a root distribution according to Hoffman and van Genuchten (1983).

The potential evapotranspiration (PET) was estimated with the Hargreaves Formula as a function of extraterrestrial radiation and daily maximum and minimum air temperature. The PET was split into potential evaporation and potential transpiration according to Beer's Law (Ritchie, 1972), which is a function of the leaf area index (LAI) and the canopy radiation extinction factor (set to 0.463).

To assess the seasonal variability of the LAI in the grassland sites (Roodt and Eichstetten), the year was divided into winter season (1st of November – 1st of March, LAI = 0.2~~ (Breuer et al., 2003)~~) and summer season (1st of May – 1st of September, LAI = 2) according to ~~(~~Breuer et al.~~,~~ (2003)~~)~~. In the transition period between the two seasons, the LAI was linearly interpolated. The interception of precipitation was considered at the grassland sites as a function of the precipitation, LAI and an empirical constant (set to 0.55 mm, which results in a maximum of 1.1 mm interception for a LAI of 2). In the scots pine forest in Hartheim, the annual average throughfall was set to be about 2/3 of the precipitation at a constant LAI of 2.8, both as reported by Jaeger and Kessler (1996). The snow module developed by Jarvis (1994) was included, where precipitation falls as snow for air temperatures <−2°C and as rain for temperatures >+2°C. Between −2°C and +2°C the percentage of snow in precipitation decreases linearly. For snow that accumulated at the soil surface, the degree-day method was applied. The required constant, which describes the amount of snowmelt during one day for each °C above zero, was set to 0.43 cm d^{-1} K^{-1}.

2.2.2. Deuterium transport

To account for the isotopic composition of the soil water, the concentration of ^{2}H~~deuterium~~ was simulated as a solute in the ~~Hydrus~~ HYDRUS model. Since the model originally was not developed to include stable isotope modelling, a modified version of HYDRUS~~Hydrus~~ was used, which was introduced by Stumpp et al. (2012) and allows for solute losses caused by evaporation. This modification prevents an accumulation of the ^{2}H~~deuterium~~ concentration at the upper boundary. The δ notation, in ‰ VSMOW of the isotopic concentration plus an offset value (to get positive values) was used for calculating the isotopic compositions and its mixing.

Isotopic enrichment due to fractionation processes during evaporation was not included in the model. This assumption was considered to have a minor impact on the simulations, because the ^{2}H~~deuterium~~-^{18}O relationship of the pore waters at the study sites were similar to the ~~global~~ local meteoric water line (L~~G~~MWL) below 30 cm soil depth, suggesting limited effects

of isotope enrichment (data not shown). Furthermore, ~~Maloszewski~~ Stumpp et al. (~~2006~~2012) found in a similar climate that the average deuterium contents in precipitation and the water outflow of a lysimeter in -~~200~~ 150 cm depth were nearly the same, concluding that fractionation due to evaporation does not play a big role in temperate climates.

Within the HYDRUS~~Hydrus~~ code, the ^{2}H~~deuterium~~ transport was calculated according to the advection-dispersion model, which is the most widely used model to predict solute transport in soils under field conditions (Vanderborght and Vereecken, 2007). The advective part of that equation is governed by the mean water flux. The dispersion term represents the hydro-chemical dispersion and the molecular diffusion. The former is a function of the longitudinal dispersivity λ [L], the water content θ [L^3 L^{-3}], and the water flux q [L T-1], while the latter is governed by the molecular diffusion coefficient in free water D_w [L^2 T-1] (2.272 *10^{-9} m^2 s^{-1} according to Mills (1973)) and a tortuosity factor τ_w [-] as defined by Millington and Quirk (1961). As ^{2}H~~deuterium~~ is part of the water molecule it can leave the soil profile via evaporation at the soil surface or via root water uptake.

The profiles ~~have been~~were discretized into 101 nodes, with higher node density at the top than at the bottom to enhance model stability. The soil profiles were discretized into two different horizons according to the soil descriptions in (Table 1). The depth of the simulation was 200 cm for Roodt and Eichstetten and 120 cm for Hartheim.

2.2.3. Initial and boundary conditions

The site-specific initial conditions were defined by a constant water content (0.2 cm^3 cm^{-3}) and a constant pore water δ^{2}H~~deuterium concentration~~, representing the weighted average concentration in precipitation (-54‰, -60‰, and -56‰ for Roodt, Eichstetten, and Hartheim, respectively). The influence of the initial conditions on the calibration can be neglected, as a spin-up period of at least ~~two years~~967 days was simulated before the start of the calibration period (Table 1). The upper boundary condition was defined by variable atmospheric conditions (Cauchy boundary condition) that govern the loss of water and deuterium caused by evapo~~transpi~~ration, the input of water due to throughfall and the accompanied flux concentrations of deuterium. Since we use a modified version of the HYDRUS code (Stumpp et al. 2012), evaporation influences only the amount of water, not its isotopic composition. The lower boundary was set to zero-gradient with free drainage of water and solutes.

2.2.4. Parameter optimization and sensitivity

Six parameters had to be optimized for each horizon of the soil profiles to simulate the water and solute transport in the unsaturated zone. On the one hand the five parameters θ_r, θ_s, α, n, K_s, describing the water retention and hydraulic conductivity characteristics in accordance to the Mualem - van Genuchten model (MVG) were determined. In addition, the longitudinal dispersivity λ, describing the dispersion of the deuterium, was subject to the optimization process. The ranges of the parameter space were based on expert knowledge and are listed in Table 2. To find the global optima of the parameter space that best simulates the observed data, the Shuffled-Complex-Evolution algorithm (SCE-UA) developed by Duan et al. (1992) was applied. The search algorithm terminates when the objective function does not improve by >0.01% within 10 evolution loops. The number of complexes used by the algorithm was defined as the number of optimizing parameters minus three, but not higher than eight or lower than three. All other parameters that govern the optimization algorithm were chosen as recommended by Duan et al. (1994). The modified Kling-Gupta-Efficiency (KGE) as defined by Kling et al. (2012) was applied as the objective function in the optimization process. The dimensionless KGE compares simulated and observed data with regard to their correlation r, their ratio of the mean values (bias ratio, β), and their ratio of the coefficient of variation (variability ratio, γ) as follows: $KGE = 1 - [(1-r)^2+(1- \beta)^2+(1- \gamma)^2]^{0.5}$. For parameter combinations that did not lead to a numerical convergence of the HYDRUS~~Hydrus~~ code, a high value of the objective function was assigned. This method, as suggested by Wöhling et al. (2008), prevents the SCE-UA algorithm from searching for an optimum in an unrealistic parameter space. A KGE was computed for each soil moisture time series at the various depths and an average KGE_θ, weighted by the number of data points for each depth was calculated to get a representative KGE for the soil moisture across the profile. Similarly, a KGE was calculated for each isotope profile and an average efficiency was derived from the mean value of all profiles (KGE_D).

The following three different inverse model approaches were tested:

1.) The isotope profile approach (IPA): Only the observed pore water isotope profiles were considered in the objective function. The MVG and dispersivity parameters were all optimized in a way to reflect the observed pore water δ^2H~~deuterium concentrations~~ in the profiles (KGE_D as objective function). The initial parameter ranges were constrained by pedotranfer functions (PTFs) using the observed soil texture (Table 1). After determining the soil texture for each horizons, the surrounding neighbours in the textural triangle were

355 determined and the corresponding MVG parameters were derived with the Rosetta PTF
356 (Schaap et al., 2001). The range of the MVG parameter values of the neighbouring textural
357 classes defined the parameter range in which the IPA was allowed to search for an optimal
358 parameter set, while the range of the dispersivity parameter was not constrained. Also an
359 variantalternative, where the parameter space of the MVG was lessnot constrained based on
360 expert knowledge (unconstrained) was tested (uIPA).

361 2.) The multi-objective approach (MOA): The measured soil moisture time series and isotope
362 profiles were used to simultaneously optimize the parameter for the water and deuterium
363 transport. Both fitting targets were equally balanced, because the KGE was calculated from
364 the average over the efficiencies of the simulated soil moisture series and the isotope profiles
365 ($KGE_{tot} = (KGE_{\theta} + KGE_{D})/2$).

366 3.) The two-step approach (2SA): The MVG parameters were optimized first by minimizing
367 the difference between observed and simulated soil moisture (KGE_{θ}). Afterwards, these MVG
368 parameters were applied in order to optimize the dispersivity parameter using the observed
369 isotope profiles (KGE_{D}).

370 In addition to the inverse model approaches, the efficiency of the simulations with parameter
371 sets derived from pedotransfer functions (PTFs) based on soil textural information of the
372 horizons were also tested to clarify the value of the pore water isotope data. The Rosetta PTF
373 (Schaap et al., 2001) was used to estimate the MVG parameters from soil textural information
374 of the horizonsand a PTF by Perfect et al. (2002) was applied for the dispersivity parameter.
375 As Rosetta does not account for the dispersivity, it was either estimated by an additional PTF
376 (Perfect et al., 2002) or again optimized using the isotope profile.

377 The sensitivities of the model to the individual parameters of the different inverse modelling
378 approaches were compared with the Regional Sensitivity Analysis (RSA) introduced by Spear
379 and Hornberger (1980) and Hornberger and Spear (1983). For each optimization approach and
380 study site, the cumulative probability distribution of the parameter set of the best performing
381 10% of model runs during the optimization process was considered for the sensitivity
382 analysis. As a sensitivity analysis, the set of model runs of the optimization process were
383 considered, whose deviation from the best run in terms of KGE was not more than 0.05 (S_{best}
384 with $KGE_i > (KGE_{best} - 0.05)$). Of this selection the 10 to 90 percentile range ($PR_{10\text{-}90}$) was
385 calculated. for each parameter as a measure for the sensitivity. Since the boundary of the
386 possible parameter space was narrowed down in the IPA via PTFs, but unconstrained in the
387 other approaches, the $PR_{10\text{-}90}$ was normalized by the range of the parameter space. Hence, the

~~PR_{10-90} represents the normalized parameter ranges between the 10th and the 90th percentile of the 10% best optimization runs as a percentage of the initial boundaries of the parameter space.~~ As the search algorithm modulation is the same for every study site and optimization approach, the PR_{10-90} allows for a comparison of the relative parameter sensitivity of the different approaches.

2.3. Water balance and transit time calculations

For each inverse modelling approach and study site, the parameter combination that resulted in the highest model efficiency ~~(Table 3) were~~ was used in a forward model approach to reveal the consequences for water balance and transit time calculations. The cumulative annual water balance from daily ~~seepage~~ recharge and evapotranspiration (ET) losses were computed over six years for each study site. To infer transit times through the soil profiles rain input was traced virtually at each study site for two events of intermediate intensities (between 8 and 13 mm day^{-1}), one that had occurred at the beginning of October (called "fall event") and one at the beginning of May (called "spring event"). We chose intermediate rain events, because such events are big enough to generate recharge and are more representative than heavier rain events, which are less likely to occur. The two different timings were considered to cover the differences of the processes over time. The sensitivities of the different approaches with regard to the water balance and transit time estimations were tested with simulations of 100 randomly chosen parameter sets from S_{best}. If the different inversely determined parameter sets lead to significant different functional responses with regard to flow and transport was tested with a one-way ANOVA and a Post-hoc analysis (Tukey's HSD). The tested variables were the mean annual ET and the median transit time, defined as the time after which half of the recharge water has passed the lower boundary of the soil profile.

3. Results

3.1. Model performance for soil moisture and pore water isotopes

The simulations with the parameter sets derived with the unconstrained isotope profile approach (uIPA) did not reproduce the soil moisture dynamics at any of the sites in a ~~satisfactory~~ realistic manner (Figure 1). The values of the KGE_θ, which did not serve as an objective function in the uIPA, ranged between -0.35 and 0.10 for the three different sites (Table 3). The models generally underestimated the water content in the upper soil layer,

whereas for Roodt and Eichstetten, the model overestimated the water content for the lower layers (at Hartheim there were no soil moisture measurements in the lower layer). For Hartheim, the high variation of the weekly measured data was not met by the simulations, but the mean of the series was reproduced. The model performance regarding the soil moisture dynamics was increased due to a constrained initial parameter space via PTFs in the IPA by 0.19, 0.61, and 0.14 for Roodt, Eichstetten and Hartheim, respectively. The IPA resulted in simulations reflecting the general pattern of the seasonal soil moisture changes. However, the other two approaches (MOA and 2SA), which included the soil moisture data in the parameterization, performed better in simulating the temporal dynamics of water contents in the soil profiles. For Roodt and Eichstetten, the KGE_θ were above 0.7 and the residuals were within the uncertainty range of the sensors ~~(±0.03 cm^3 cm^{-3})~~ except for dry periods in Eichstetten. For Roodt, where the observed soil moisture time series are averages of three sensors per depth, the deviation of the three sensors from their average value was higher (0.03 to 0.08 cm^3 cm^{-3}) than the residuals of the simulations of MOA and 2SA. The model efficiency for soil moisture dynamics at Hartheim is lower than for the other study sites (KGE_θ 0.20 and 0.42 for the MOA and the 2SA, respectively). The modelled soil moisture data with the best parameter set of MOA does not reflect the temporal variability of the observed data, but the mean values are reproduced. With the parameter set resulting from the 2SA, the dynamics, as represented by the coefficient of variation in the KGE, are better simulated, but the correlation between observed and simulated data is lower.

For the pore water ~~deuterium concentrations~~isotope profiles, the best fits with KGE_D between 0.72 and 0.86 were achieved with the parameters derived from uIPA (Figure 2 and Table 3). Constraining the parameter space (IPA) led to a decrease of the KGE_D by 0.07 to 0.11. Including soil moisture data into the calibration (MOA) reduced the KGE_D moderately to values between 0.67 and 0.81. Parameters derived with the 2SA resulted in slightly lower model efficiency at Roodt and Eichstetten with a KGE_D of 0.62 and 0.79, respectively. For Hartheim, the 2SA resulted in the lowest KGE_D of 0.40. The fit between simulated and observed pore water isotope concentrations is not equally good for all the sampling times at the same sampling site. For Roodt, the isotope profile from October was better simulated than the profile sampled in March. While the peak of isotopically enriched water from summer precipitation in 30 to 50 cm soil depth is well simulated in the October profile, there is a higher vertical variability in the simulated profile than in the observations. For Eichstetten, the isotope profile in November was reproduced more closely than the ones taken in January and March. Temporal dynamics of the model fit are less pronounced for the site in Hartheim,

where the vertical variability across the soil profile is generally lower than at the other two study sites. Estimating the MVG parameter with the Rosetta pedotransfer function (PTF) (Schaap et al., 2001) via textural information ~~at Roodt and Eichstetten~~, did not result in a proper representation of the soil moisture dynamics (~~KGE_θ of 0.03 and 0.27, respectively~~Table 3). Using the texturally dependent PTF for the dispersivity parameters (Perfect et al., 2002) in combination with the MVG parameters from the Rosetta PTF failed to simulate the measured pore water isotope concentrations in Roodt (KGE_D = -0.17), while the result for Eichstetten (KGE_D = 0.43) and Hartheim (KGE_D = 0.44) was better ~~(KGE_D = 0.43)~~. ~~An inverse estimation of the dispersivity parameter with KGE_D as the objective function while using the MVG of the Rosetta PTF resulted in higher model efficiencies as with the dispersivity parameters derived from the PTF of Perfect et al. (2002). However, the efficiencies were much lower than for the three presented inverse model approaches, with KGE_D of 0.18 and 0.59 for Roodt and Eichstetten, respectively.~~

3.2. Parameter sensitivity

The sensitivity analysis showed that the range of the parameters ($PR_{10\text{-}90}$) of the set of the best performing parameter combinations S_{best} vary strongly between the different inverse modelling approaches and study sites. While the parameter range is low for the MOA at Eichstetten, the MOA results in higher parameter ranges for Roodt and intermediate ranges for Hartheim (Figure 3). The 2SA results in high $PR_{10\text{-}90}$ values for Eichstetten and Hartheim, but for Roodt, the 2SA results in low ranges. The uIPA and IPA give small to intermediate $PR_{10\text{-}90}$ values for all three sites. Generally, the parameters of the upper soil horizons at Roodt and Eichstetten are less sensitive – independent of the inverse model approach. This pattern is less pronounced for Hartheim, where only the 2SA shows a distinct lower sensitivity for the second horizon. Lowest sensitivities for all sites and approaches can be detected for K_s, θ_r, and θ_s, while the parameters λ, n, and α are better identifiable.

~~The sensitivity analysis based on the 10-90 percentile ranges of the 10% best parameter sets showed that the uIPA results in least identifiable parameters for Roodt and Eichstetten, while the MOA gives the best results regarding the parameter sensitivity (Figure 3). The $PR_{10\text{-}90}$ values of the uIPA are at least 4 and 2 times higher than for the MOA at Roodt and Eichstetten, respectively. The sensitivities of the parameters from the 2SA take an intermediate position, where the K_s parameters show lowest sensitivities ($PR_{10\text{-}90}$ 10% to 16% for Roodt and Eichstetten, respectively). For Hartheim the pattern in the relationship between optimization approach and parameter sensitivities is less pronounced and the MOA does not~~

~~result in the best identifiable parameters. At this site only, the 2SA results in a substantial difference in sensitivities between the upper and lower layer: MVG parameters of horizon 1 (except for α) have 3 to 23 times lower PR_{10-90} values than the parameters of horizon 2. In general, K_s shows the lowest sensitivities for all sites and optimization approaches (more than half of the PR_{10-90} values are above 5%, respectively), while θ_r, and θ_s are of immediate sensitivities (1/3 of the PR_{10-90} are >5%) and both α and n are usually well identified (1/4 and 1/5 of the PR_{10-90} values are >5 %, respectively).~~

The water retention curves and the unsaturated hydraulic conductivity for Roodt and Eichstetten are similar for the MOA and the 2SA, while the IPA and especially the uIPA yielded parameter combinations that result in rather different retention curves (Figure 4 and Table 4). This pattern is less pronounced for the different inverse modelling approaches for Hartheim. For Roodt, the dispersivity is higher in the upper layer, while it is higher in the lower layer for Eichstetten and Hartheim using the MOA and 2SA (Table 4).

3.3. Consequences for the water balance and water transit times

Magnitudes of site-specific water balance components derived with the MOA and 2SA are generally of similar range (Figure 5). The water balance components derived with the uIPA deviate from the other inverse modelling approaches resulting in high ~~seepage~~ recharge fluxes and low ~~evapotranspiration~~ ET for Roodt and ~~Hartheim~~Eichstetten. These high ~~seepage~~ recharge rates, which are twice as high as the ET for Eichstetten, are due to the low saturated water content and high hydraulic conductivities in the upper soil horizon estimated by the uIPA. The water balance simulated with the uIPA for Eichstetten is not realistic, since the annual ET is reported to be about 80% of the precipitation (ET/P = 0.8) in this region (upper Rhine Valley) (Wenzel et al., 1997). In contrast, the IPA, MOA and 2SA result in an ET/P between 0.77 and 0.82 for three of the four simulated years. For Hartheim the simulated ET/P ratios are with 0.63 to 0.85 in a similar range as derived from latent heat flux estimates (ET/P = 0.71 to 0.88) for the years 2000 and 2001 (Imbery, 2005). The statistical analysis showed that the inverse model approaches resulted in significantly different mean annual ET estimates when considering the different parameter combinations of the set S_{best} (Table 5).

The fact that parameters derived with the different optimization approaches differ less for Roodt and Hartheim than for Eichstetten is also reflected in the results of the transit time estimations. Cumulative breakthrough curves of the traced event waters leaving the soil profile at the lower boundary were determined for two events (Fig. 6). Figure 6 does not only visualize the timing and amount of event water in the ~~seepage~~ recharge flux, but also the

fraction of ~~seepage~~ recharge water to ET (i.e. difference to unity). There are pronounced seasonal effects with at least four times higher ~~seepage~~recharge-ET ratios for the rain event in fall than for the spring event. In general, precipitation in fall is more likely to leave the soil via ~~seepage~~ recharge and to do so after shorter transit times. ~~Big~~ Pronounced differences between the approaches were found for Eichstetten, where the uIPA resulted due to the low θ_s in transit times ~~are~~ that were ~~twice as long for~~ two times shorter as the IPA, MOA and the 2SA ~~than for the uIPA, respectively~~ (Table 5). The mean transit times (MTT) simulated with 100 randomly chosen parameter combinations from S_{best}, are statistically significant different among the inverse model approaches for Eichstetten. For Roodt, transit times of the IPA and uIPA were about ~~double~~ twice as long as for the MOA and 2SA and the latter two approaches did not differ significantly in terms of MTT. For Hartheim, the uIPA and the MOA did not differ significantly with regard to the MTT, while the others did.

4. Discussion

4.1. Parameter adequacy

The MOA shows highest overall parameter adequacy when challenging the results of the conducted model calibrations in accordance to Gupta et al. (2005) with regard to: i) the fit between observed and simulated data, ii) accuracy of the parameter sets, and iii) consistency of the model behaviour. The MOA outcompetes the other inverse model approaches with respect to the overall efficiency (KGE_{tot}) of the simulation of both the soil moisture dynamics and pore water isotope concentrations (Table 3), while the sensitivity of the parameters derived with the MOA is more variable. The model results regarding the water balance and transit times are similar for the 2SA and IPA and generally of the same magnitude of measured water balance estimations. ~~: parameter identifiability is the highest and the simulated water balance components and transit times are in line with the understanding of the processes at the study sites.~~ The 2SA gave satisfactory results in the model efficiencies and model consistencies, but also showed variable results ~~weaknesses~~ regarding the identifiability of the parameters due to the fact that five MVG parameters for two horizons were optimized with just one objective function (KGE_θ) in the first step (see MVG for Eichstetten and Hartheim in Fig.3). The uIPA, where also just one objective function was applied (KGE_D), showed problems with respect to the ~~identifiability~~ parameter identifiability in the upper horizons as well as low model performance and realism. The identifiability of the IPA appears to be well in Figure 3, but caution has to be paid since some parameters moved to

the boundaries of the parameter space set by the Rosetta PTF, resulting in little or no changes within the best performing ~~10% of the~~ optimization runs (e.g. for Roodt 7 out of the 12 parameters reached boundaries). All parameters that moved to the boundaries during the optimization with the IPA are indicated with a star in Table 4. Despite this limitation, the IPA reveals that the information about soil texture to limit the possible parameter range helps to find an overall more realistic parameter set. Constraining the possible parameter space of the MVG parameters resulted in increased KGE_{tot}, while the objective function of the IPA (KGE_D) resulted in slightly lower values.

The inadequate representation of the soil moisture dynamics using the hydraulic properties~~soil physical parameters~~ derived with the Rosetta PTF (Table 3) shows that site-specific hydrological characteristics can hardly be reflected via ~~solely~~ textural information alone. This limited accuracy of PTFs which use only soil texture was also found in other studies as reviewed by Vereecken et al. (2010)~~. Nevertheless~~, indicating that soil structure has to be taken into account. This is especially true for Roodt, where a high rock content influences the water flow. Therefore, the application of the PTF results in a better simulation for Eichstetten and Hartheim than for Roodt, which indicates that the flow in the first two study sites is more homogenous. At Roodt, the PTF fails to represent the water flow (KGE_θ = -0.17), but the MOA and 2SA result in satisfactory simulations, showing that the inverse estimated parameters are effective parameters that hold information of non-heterogeneous flow that cannot be represented in the model. ~~The fact that the soil moisture simulations as well as the pore water isotope simulations with parameters derived via the PTF are better for Eichstetten, where a more homogenous soil prevails than in Roodt, supports the idea that the application of PTFs require homogenous flow through the soil matrix.~~ As an example, measurements of K_s on soil cores taken in the catchment of the study site in Roodt showed high variability of the hydraulic conductivity with values ranging between 29 and 2306 cm day^{-1} across the soil profile. The inversely estimated K_s-values for Roodt lay within the range of these measurements. Further estimations of the MVG parameters on soil cores taken in the upper horizon in the study area at Roodt showed similar ranges as the parameter sets derived via inverse modelling. Exceptions are the parameter n, which has higher values for the uIPA and IPA than the laboratory measurements, and the θ_s, which is generally lower for the inverse optimization compared to the measurements, which could reflect the influence of the rock content. The deviation between the inverse estimations and laboratory measurements could also be due to the lack of high volumetric water contents in the soil moisture data and the fact that the soil moisture sensors are not calibrated. For the other study sites, no laboratory

measurements on soil cores are available, but infiltration experiments with Uranin showed that that water introduced during fall events percolated down to 140 cm during one year (Koeniger, 2003) at Hartheim, which is well reproduced with the MOA and slightly overestimated by the other approaches (Table 5). Furthermore, infiltration measurements at Hartheim with a double ring revealed a high variability of the saturated hydraulic conductivity (1 – 800 cm day^{-1}) in the topsoil, and the inversely estimated K_s parameters are within this range.

In general the KGE_{tot} was lower in the approaches that made use of PTF than for the MOA and the 2SA, which shows the advantage of including both, the hydrometric and hydrochemical data in inverse modelling ~~in~~ for effectively and site specifically optimizing the model parameters. Our findings support the acknowledged fact that PTFs have a limited transferability from the region and scale they were developed, since they do not account for the pore structure (Pachepsky et al., 2006). Even though the soil is not a homogenous porous medium as assumed for the applied Richards equation, our simulations of water flow and isotope transport on daily resolution over several years seems to capture the hydrological processes of percolation, ~~evapotranspiration~~ ET and dispersion of pore waters reasonably well in terms of soil moisture dynamics and isotope composition of the pore waters. Highest deviations of the modelled soil moisture dynamics from the observed data are found during dry periods. The overestimation of the water content in these cases is likely caused by the simplified root distribution and water uptake model. The highest deviations of the modelled pore water stable isotope composition from the observed isotope profiles are found for the sampling in January and March, which could be caused by an insufficient representation of the snow melt processes or transpiration. Also preferential flows, which were shown to occur mainly during the wet season after snow melt (Gazis and Feng, 2004; Mueller et al., 2014) might cause bigger differences between observed and simulated isotope profiles during winter times. Thus, the number of considered isotope profiles and their sampling timing can have an important impact on the inverse model approaches. Generally, it is preferable to have several pore water stable isotope profiles taken during different seasons and hydrological states.

4.2. Dispersivity parameter estimation

An increase of the dispersivity parameter with depth and length, as found in several core-, column-, and lysimeter experiments (summarized by Pachepsky et al. (2000) and Vanderborght and Vereecken (2007)) was only found for Eichstetten. For Roodt and Hartheim, the dispersivity was higher in the upper horizon. However, the scale dependency of

the dispersivity is generally reported to be less pronounced or non-existent for the field scale experiments and longer travel distances (Vanderborght and Vereecken, 2007). The estimated values for the dispersivity parameters are mostly within the range (0.8 – 20 cm) as reported in a review by Vanderborght and Vereecken (2007) for the field scale~~. However, the results of the dispersivity parameters derived by the MOA and 2SA for Roodt and Eichstetten (0.12 – 9 cm), which can be seen as most reliable simulations, are at the lower end of the data presented in the review by Vanderborght and Vereecken (2007)~~ and lysimeter studies by Stumpp et al. (2009a~~) and Stumpp et al. (~~2012). As the dispersivity parameter was shown to be scale dependent (Vanderborght and Vereecken, 2007), the presented methodology provides the opportunity to optimize parameters for each soil horizon, in contrast to soil column or lysimeter studies, where the dispersivity parameter is integrated over the entire soil profile (Inoue et al., 2000; Stumpp et al., 2012). In addition, only 1 to 2 sampling campaigns are necessary to get the additional information for water and solute transport. The high variability of the dispersivity between the sites and horizons in our study and reported in other studies (Vanderborght and Vereecken, 2007) and the limited model efficiencies when PTFs were applied emphasize the importance to consider ~~this solute transport parameter~~ the dispersivity in the parameterization of soil physical models. ~~Thus, a~~A field scale representation of the dispersion processes cannot ~~therefore not~~ be assumed for a certain soil texture by a PTF, but should rather ~~must~~ be derived for the particular field site. ~~The fact that the soil moisture sampling at Hartheim was constrained to the upper soil layer but pore water isotope data reached to depth of 120 cm explains the large differences between MOA and 2SA at that study site. The MVG parameters of the lower layer are in this case non-identifiable. Nevertheless, the MOA results in reasonable simulations of the soil moisture and the isotope profiles in Hartheim.~~ Since the efficiency of the pore water isotope simulations is beside the MVG and dispersivity parameter highly dependent on the isotopic signal of the rainwater, a sufficiently long input time series is crucial in order to ensure that the initial pore water has been renewed over the simulation period to minimize the influence of the initial conditions. In our case, this is given since the spin-up periods (Table 1) are generally longer than the estimated transit times (Fig. 6).

4.3. Advantages of multi-objective approaches

Our comparative study supports the findings by others that the more data types are taken into account during the calibration process, the lower is the model's performance with respect to different specific objective functions. For catchment models it has been shown that including

651 stream water chloride (Kuczera and Mroczkowski, 1998) or isotope concentrations (Fenicia et
652 al., 2008; Hartmann et al., 2012) in the optimization process reduced stream discharge
653 simulation efficiency, but increased model realism and parameter identifiability. On a
654 different scale, a similar effect was reported for soil physical models, as shown in
655 comparative studies, where soil moisture data from soil cores were combined with pressure
656 heads (Zhang et al., 2003; Vrugt and Bouten, 2002) or with leachate volume of lysimeters
657 (Mertens et al., 2006) to increase identifiability. Our study is in line with these findings, but
658 expanded the comparison to the field scale and included hydrochemical data. The
659 simultaneous optimization outcompeted in two of three cases the two-step optimization with
660 regard to identifiability (as also found by Mishra and Parker, 1989), while providing similar
661 overall performance as the 2SA. The MOA has the advantage that the MVG parameters are
662 additionally constrained by the percolation velocity in the advection-dispersion function used
663 to simulate the isotope profile, and not just by the soil moisture dynamics, as for the 2SA.
664 Another advantage is the lower time requirement for the calibration using MOA, because the
665 parameterization is done in one and not in two subsequent steps. Considering these
666 advantages, with a performance that is as good as for the 2SA, and much better than the IPA
667 and uIPA, the MOA represents the best inverse model approach. These findings are in line
668 with Mishra and Parker (1989), who also found the simultaneous estimation of hydraulic and
669 transport properties to be better than the sequential inversion of first hydraulic properties from
670 water content and matric pressure head data, followed by inversion of transport properties
671 from concentration data. Inoue et al. (2000) also showed a successful application of the
672 simultaneous optimization of soil hydraulic and solute transport parameters, but did not
673 compare the performance with a two-step optimization. In accordance to our findings that the
674 KGE_θ was only slightly lower for the MOA than for the 2SA (Table 3), Abbasi et al. (2003a)
675 found a better performance for the simulation of the soil moisture data when the two-step
676 approach was applied. However, with respect to drainage rates and concentrations, the
677 simultaneous optimization of the water flow and solute transport parameters resulted in as
678 good model performances as the sequential approach (Abbasi et al.; 2003a, Jacques et al.
679 2002). In our study, we aimed to represent the water flow and isotope transport on the pedon
680 scale as complex as needed, but as simple as possible. Therefore, processes like preferential
681 flow, hysteresis or mobile-immobile interactions in the soil were not considered. Including
682 these processes in the model would cause a need for more parameters, which is likely to result
683 in lower identifiability. However, even in this case the additional isotope data may help to
684 better constrain the parameters.

4.4. Transit time estimations

There is an additional benefit in taking isotope data into consideration in soil physical models with respect to the possibility of tracing the water movement through the soil. The fact that the pore water isotope data allows us to determine the dispersion of the water during the percolation processes provides the opportunity to apply particle tracking of the precipitation water, which would not be possible with an inverse model approach limited to hydrometric data. ~~As a consequence, the presented inverse model approaches that include the water's isotopic signal on the field scale outcompete the usually applied parameterization of soil physical models that consider only hydrometric data.~~ By simulating the isotope transport in the unsaturated zone, not only the response time, but also the transit time of the water can be predicted, which provides additional valuable information for a better understanding of the hydrological processes in the subsurface.

The simulated transit time distributions reveal that the water transport can differ by several weeks to months, depending on the inverse modelling approach, while the water balance estimations seem to be less sensitive to the method used to derived the parameter sets (except for the uIPA). Besides the timing of the tracer breakthrough, also the amount of recharge is sensitive to the estimated parameter set as shown in the deviation between maximum actual cumulative recharge and total possible recharge (= 1 in the cumulative density functions in Fig. 6). Thus, our study showed that the parameter estimation for soil physical models is more crucial for transit time modelling than for water balance calculations.

The presented inverse model approaches ~~is~~ are limited to environments where a seasonal variation in the isotopic composition of precipitation exists and soil evaporation and thus isotopic fractionation processes play a minor role. However, isotope fractionation processes due to evaporation could also be included in a Richards based model. The presented inverse model approaches including the estimation of the dispersivity parameter at the field scale will be beneficial for studies dealing with pollutant and nutrient transport through the soil.

5. Conclusion

We conclude that the information gained by the snap shot sampling of soil water isotope profiles allows for a more realistic parameterization of soil physical models. Our study showed the strength of pore water isotope information as fitting target for the parameterization of soil physical models. Stable water isotope profiles as the only optimization target (uIPA) do not provide sufficient information to derive hydraulic

properties~~soil physical parameters~~ that can reflect the soil moisture dynamics, but constraining the possible parameter space of the MVG parameters with information about the soil texture (IPA) helps to increase model realism. Continuous measurements of the water content or the matric potential seem to be still beneficial for understanding the water movement within the soil profile. Regarding water balance and transit time simulations, the uIPA and IPA have to be applied with caution and model realism has to be tested, for example by field measurements of ET and/or soil storage changes. Since the identifiability is higher for the MOA than for the 2SA in two of three considered cases, while the model performance and realism are similar, the combination of pore water isotope profiles and soil moisture time series as parallel optimization targets (MOA) result in the most adequate parameter representation. Parameters derived via PTFs did not lead to ~~satisfying~~ realistic simulations.

In general, the consideration of the isotopic signal enables an estimation of the dispersion of the water during the percolation through the soil. As such, tracking of the infiltrated water is possible, which gives insights into the transit times - and not just the response times - of the soil water on the field scale. Hence, isotope profiles in combination with soil moisture time series feature the opportunity to derive time-varying, site-specific transit time distributions of the vadose zone via soil physical models. Although the information is limited to point measurements, a better knowledge of the water velocities and mixing processes will help to benchmark conceptual catchment models. It seems even possible to realistically estimate soil hydraulic parameters from pore water stable isotope profiles alone. This will reduce the time and effort for long-term soil water content measurements significantly, since only one to two sampling campaigns to extract soil samples are necessary. However, longer time series of rainfall and isotopic composition are crucial for the presented approaches.

Tackling the limitations of the here presented study by including preferential flow and isotopic fractionation due to evaporation would open up additional avenues such as estimating the impact of heavy precipitation events and resulting preferential flow on the water and solute transport or differentiating between evaporated and transpired soil water. Overall, we expect the more realistic parameterization of soil physical models based on the inclusion of pore water isotope data to improve the assessment of groundwater pollution by water soluble nutrients, pesticides or contaminants.

Author contribution

M.S. performed the simulations and prepared the manuscript with contributions from all co-authors. T.H.M.V. provided the data for Eichstetten. T.B. and M.W. designed the experiment. All authors contributed to the writing of the manuscript with M.S. taking the lead.

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Figures

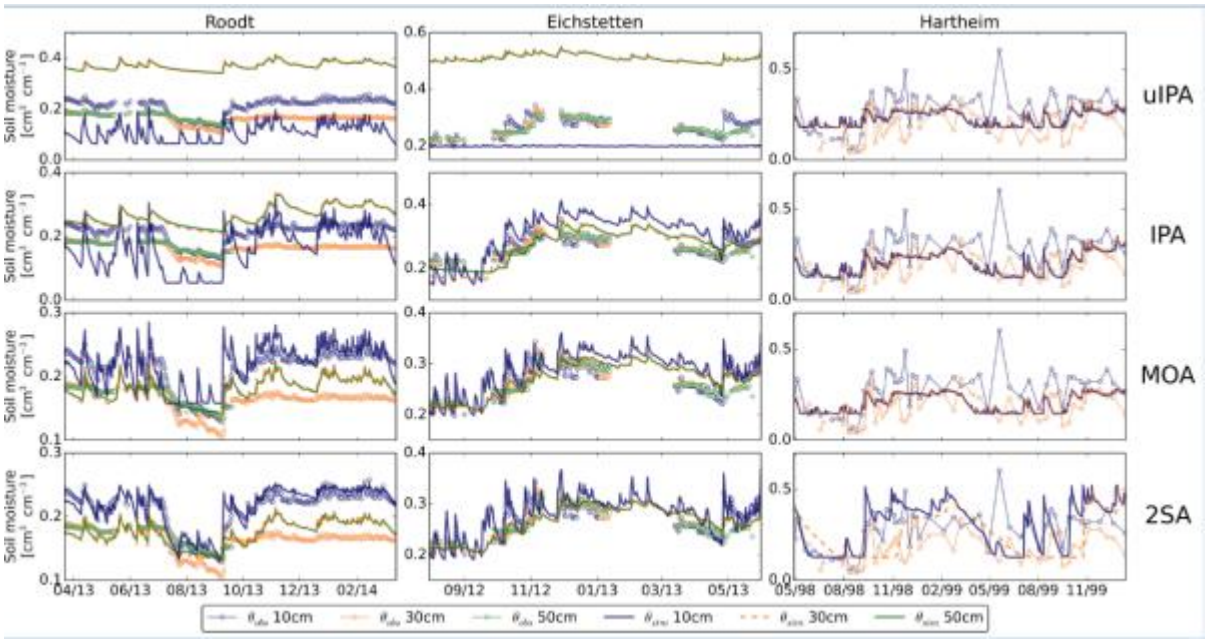

Figure 1 Observed soil moisture (circles) at each study site and the corresponding simulated soil moisture (lines), modelled with the best parameter set derived from the three different inverse model approaches. Two or three observed soil moisture time series are shown. uIPA: Unconstrained isotope profile approach; IPA: Isotope profile approach; MOA: Multi-objective approach; 2SA: Two-step approach.

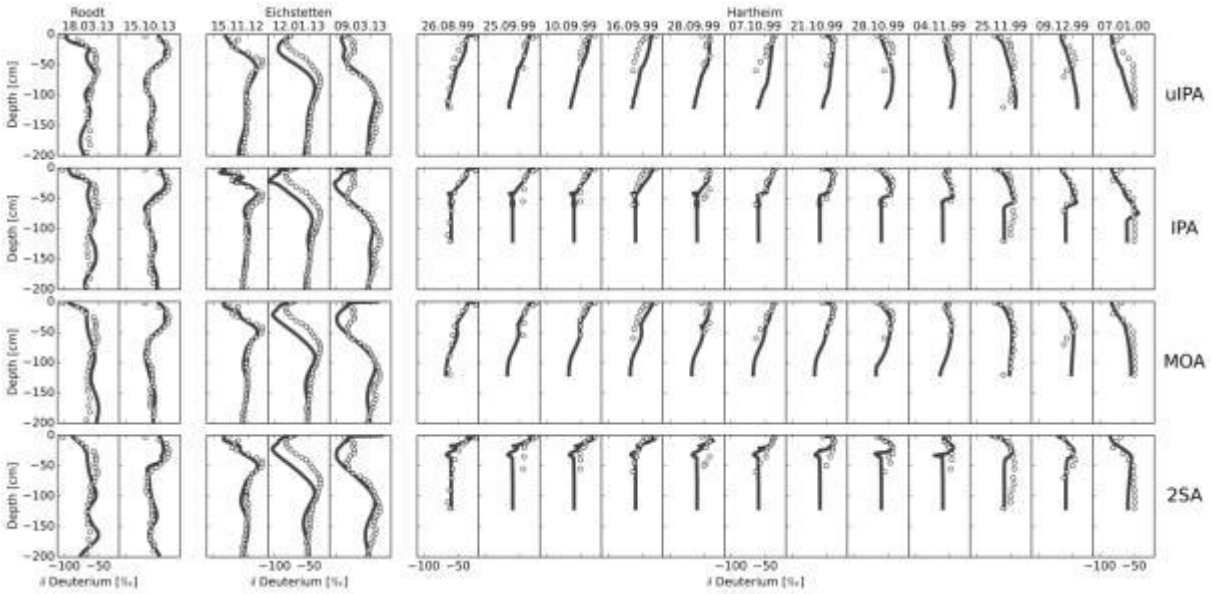

Figure 2 Observed (circles) and simulated (lines) pore water deuterium concentrations at each study site and at various dates. Simulations done with the best parameter set derived from the three different inverse model approaches. Axes scaling kept constant for each subplot.

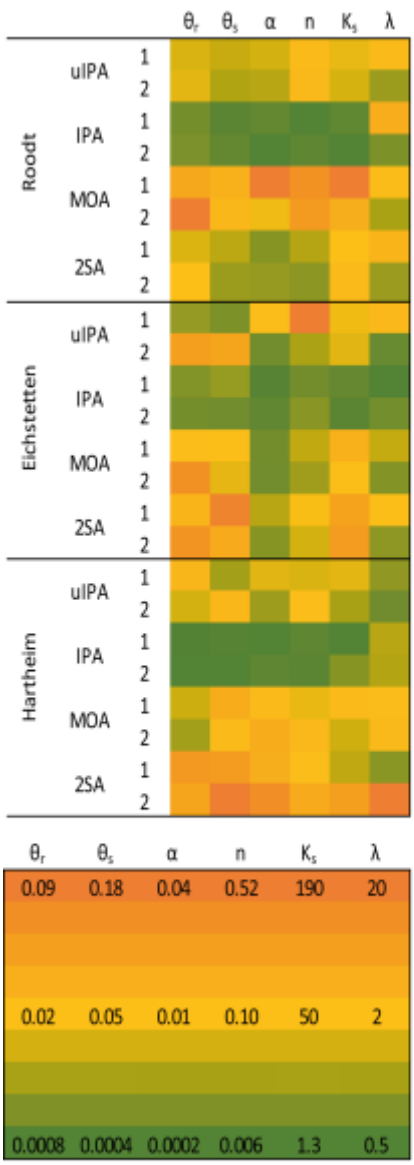

Figure 3 Parameter identifiability of each parameter calibrated at each site with the different inverse model approaches (uIPA, IPA, MOA, 2SA) for the upper (1) and lower (2) soil horizon. Colour indicates the ~~normalized~~ parameter ranges between the 10th and the 90th percentile of the ~~10% best optimization runs~~of the parameter combinations of the set S_{best} for each approach and study site. ~~as a percentage of the initial boundaries of the parameter space~~. Green indicates a small range, yellow medium and orange represents a high range.

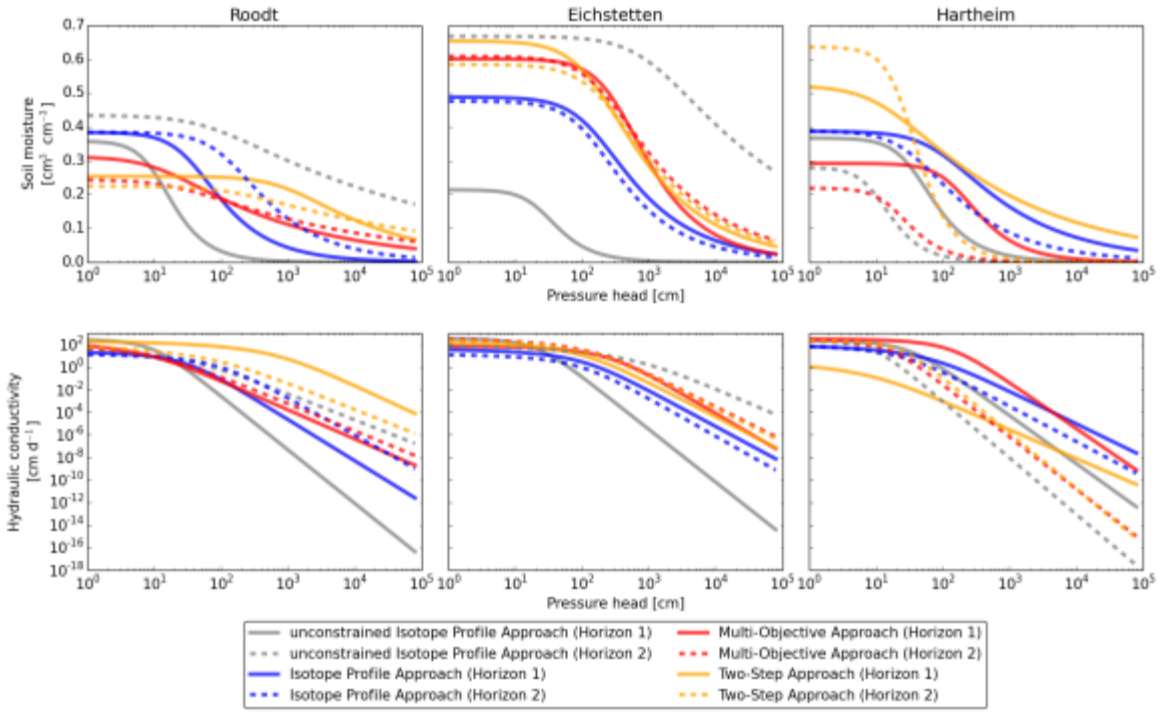

Figure 4 The water retention and the hydraulic conductivity functions for the parameter sets of the upper and lower soil horizons (continuous and dashed line, respectively), that resulted in the best model performance after calibrating with the three different inverse modelling approaches for each study site. Note that with respect to these characteristic curves the 3 calibration approaches are based on only isotope data (uIPA), a mix of isotope data and soil texture data (IPA), a mix of isotope and soil moisture data (MOA) and only soil moisture data (2SA).

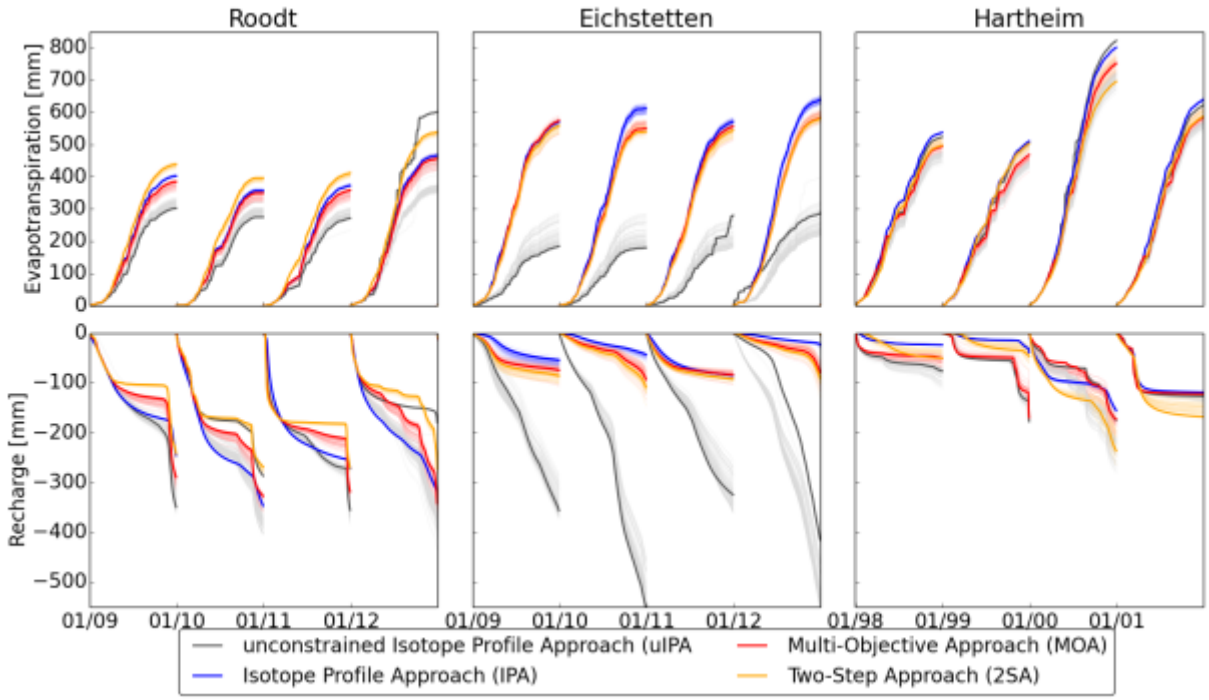

Figure 5 Annual simulated cumulative actual evapotranspiration (first row) and cumulative ~~seepage~~ recharge through the -200 cm (Roodt and Eichstetten) and -120 cm (Hartheim) depth plane (~~dashed lines~~lower row) ~~and actual evapotranspiration (continuous lines). simulated with the best parameter sets optimized with the different inverse modelling approaches for each site.~~ Solid lines show simulations with the parameter sets that performed best during the different inverse modelling approaches at each study site and the thin transparent lines represent simulations with 100 randomly chosen parameter combinations of the set S_{best}.

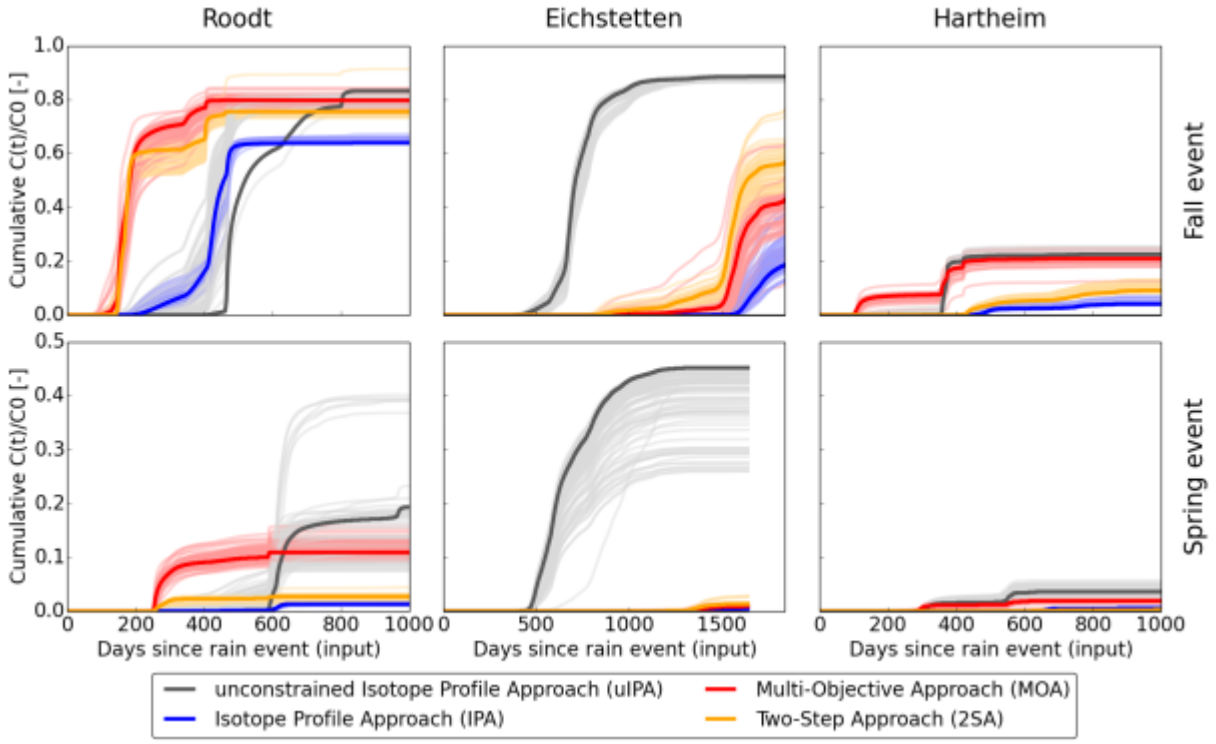

Figure 6 Cumulative transit time distribution of rainwater infiltrated during an event in fall (~~continuous line~~first row) and spring (~~dotted line~~second row) in the ~~seepage~~ recharge flux through the -200 cm (Roodt and Eichstetten) and -120 cm (Hartheim) depth plane. Solid lines show ~~S~~simulations ~~were done~~ with the parameter sets that performed best during the different inverse modelling approaches (colours) at each study site and the thin transparent lines represent simulations with 100 randomly chosen parameter combinations of the set S_{best}.

Tables

Table 1 Environmental characteristics of the three study sites and the available data for the fitting targets for the inverse modelling.

		Roodt	Eichstetten	Hartheim
Location		49°82' N; 5°83' E	48°05’ N; 7°42’ E	47°56' N; 7°36' E
Elevation m a.s.l.		470 m	310 m	200 m
Geology		Devonian Schist[a)]	Pleistocene aeolian loess	Fluvial Gravel[d)]
Soil type		Cambisol	Luvisol	Calcaric Regosol[d)]
Soil depth	Horizon 1	0 - 25 cm	0 - 25 cm	0 - 40 cm[d)]
	Horizon 2	>25 cm	>25 cm	>40 cm[d)]
Soil texture	Horizon 1	loam	~~slightly clayey~~ silt	s~~S~~ilty ~~sand~~[d] loam[d)]
	Horizon 2	clayey loam	~~slightly clayey~~ silt	fluvial gravel and coarse sand[d)]
Mean annual temperature [°C]		8.3[b)]	11	9.8[e)]
Mean annual precipitation [mm]		845[b)]	900	667[e)]
Land use		Grassland	Grassland	Pinus sylvestris (Scots pine)[e)]
Maximum rooting depth [cm]		-20	-30	-40
Soil moisture data	Sampling period	Daily (22 Mar 2013 – 15 Mar 2014)	Daily (31 Jul 2012 – 31 May 2013	Biweekly (29 Apr 1998 – 13 Jan 2000)
	Sampling depth [cm]	-10, -30, -50 (each as average of three replicates)	-5, -10, -20, -30, –40, -50, -60	-2, -10 ,-30
	Sampling method	5TE sensors (Decagon)	5TE sensors (Decagon)	Gravimetric with soil cores[f)]

Isotope profiles sampling (first and last sampling date)	2 (18 Mar 2013 – 15 Oct 2013)	3 (15 Nov 2012 – 09 Mar 2013)	16 (26 Aug 1999 – 07 Jan 2000)
Pore water isotope analysis	Equilibrium method[c)]	Equilibrium method[c)]	azeotropic distillation with toluol and mass spectrometer[g)]
Model period (included spin-up period)	01 Jan 2008 – 31 Dec 2013 (1903 days)	01 Jan 2008 – 04 Nov 2013 (1780 days)	01 Jan 1997 – 31 Dec 2002 (967 days)

[a)] (Lorz et al., 2011); [b)] (Pfister et al., 2005); [c)] Wassenaar et al. (2008); [d)] (Schäfer, 1977); [e)] (Mayer et al., 2005); [f)] (Koeniger, 2003); [g)] (Revesz and Woods, 1990)

Table 2 Boundaries of the parameter space for the unconstrained inverse model approaches (uIPA, MOA, 2SA).

Parameter	Lower boundary	Upper boundary
Residual volumetric water content, θ_r [cm^3 cm^{-3}]	0	0.2
Saturated volumetric water content, θ_s [cm^3 cm^{-3}]	0.2	0.7
inverse of the capillary fringe thickness, α [cm^{-1}]	0.001	0.1
MVG shape parameter, n [-]	1.1	2.5
Saturated hydraulic conductivity, K_s [cm day^{-1}]	10	400
Longitudinal dispersivity, λ [cm]	0	30

Table 3 Performance of the pedotransferfunctions (PTF) and the different inverse model approaches (uIPA, IPA, MOA, 2SA) regarding the soil moisture (KGE_θ) and isotope (KGE_D) data and the average of this both efficiency measure (KGE_{tot}) for the three study sites. (Perfect fit would result in a Kling-Gupta-Efficiency (KGE) of 1.)

	Roodt			Eichstetten			Hartheim		
	KGE_θ	KGE_D	KGE_{tot}	KGE_θ	KGE_D	KGE_{tot}	KGE_θ	KGE_D	KGE_{tot}
PTF	-0.17	0.48	0.15	0.17	0.43	0.40	0.37	0.44	0.41
uIPA	-0.35	0.83	0.24	0.37	0.86	0.31	0.10	0.72	0.41
IPA	-0.15	0.72	0.28	0.37	0.80	0.58	0.24	0.65	0.45
MOA	0.70	0.69	0.70	0.79	0.82	0.80	0.20	0.67	0.44
2SA	0.80	0.62	0.71	0.80	0.79	0.80	0.43	0.40	0.41

Table 4 Best performing parameter sets of the different optimization approaches for the three different study sites. * indicate parameter that reached the initial boundaries of the parameter space in the IPA.

Study site	Optimization approach	Horizon	θ_r	θ_s	α	n	K_s	λ
Roodt	PTF	1	0.078	0.43	0.036	1.56	25	4.6
		2	0.095	0.41	0.019	1.31	6	8.1
	uIPA	1	0.065	0.358	0.089	2.10	295	4.3
		2	0.072	0.434	0.017	1.13	238	1.0
	IPA	1	0.044	0.384*	0.027*	1.66*	24	23.2
		2	0.074	0.384*	0.008*	1.52*	15*	0.4
	MOA	1	0.115	0.312	0.081	1.23	378	2.7
		2	0.014	0.244	0.047	1.17	301	1.0
	2SA	1	0.052	0.254	0.001	1.30	242	9.0
		2	0.021	0.225	0.007	1.14	242	0.1
Eichstetten	PTF	1	0.034	0.46	0.016	1.37	6	5.6
		2	0.034	0.46	0.016	1.37	6	5.6
	uIPA	1	0.197	0.214	0.040	2.07	355	7.1
		2	0.026	0.668	0.001	1.21	129	4.2
	IPA	1	0.038	0.488*	0.007*	1.48*	40	0.1
		2	0.067	0.476	0.008	1.54	14	2.5
	MOA	1	0.122	0.601	0.003	1.59	76	0.7
		2	0.012	0.609	0.005	1.38	394	1.8
	2SA	1	0.076	0.654	0.007	1.42	185	0.5
		2	0.011	0.585	0.005	1.39	306	1.8
Hartheim	PTF	1	0.067	0.450	0.02	1.41	11	5.6
		2	0.045	0.430	0.145	2.68	713	0.8

uIPA	1	0.179	0.367	0.026	1.90	237	8.0
	2	0.045	0.280	0.095	2.21	243	0.0
IPA	1	0.059	0.387*	0.011	1.35	104*	8.2
	2	0.041	0.388	0.026*	1.45	104*	0.2
MOA	1	0.141	0.292	0.006	1.83	308	9.1
	2	0.028	0.219	0.052	2.06	228	15.2
2SA	1	0.004	0.522	0.078	1.22	6	1.8
	2	0.104	0.636	0.036	2.17	223	29.2

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Table 5 The median transit time (MTT) of the two rain events in fall and spring, whose water was traced virtually through the vadose zone and the modelled average annual evapotranspiration (ET). The values are results for the best performing parameter set and the given ranges are the standard deviation of the randomly sampled 100 parameter combinations of the set S_{best}.

Site	Model approach	MTT 'Fall event' [days]	MTT 'Spring event' [days]	Mean annual ET [mm]
Roodt	uIPA	495 ± 22	626 ± 14	362 ± 10
	IPA	425 ± 6	613 ± 3	399 ± 2
	MOA	173 ± 7	275 ± 10	387 ± 8
	2SA	172 ± 1	281 ± 3	446 ± 3
Eichstetten	uIPA	697 ± 14	624 ± 45	232 ± 28
	IPA	1685 ± 14	1503 ± 11	598 ± 7
	MOA	1579 ± 24	1399 ± 24	565 ± 7
	2SA	1543 ± 5	1372 ± 5	556 ± 7
Hartheim	uIPA	370 ± 2	540 ± 4	617 ± 9
	IPA	510 ± 13	672 ± 40	621 ± 1
	MOA	359 ± 7	317 ± 74	574 ± 8
	2SA	545 ± 21	697 ± 5	570 ± 12