Rebuttal to interactive comment on hess-2016-374-RC1

1) Generally speaking, the structure of this manuscript is more like a hydrogeological survey report or groundwater resource summary, not a research article. Why did you do this study? What scientific questions are answered in this paper? The author listed three aims in the introduction part, but it seems the authors are trying to address so many issues in one manuscript, and bring difficulties for readers to follow up. The first aim is obviously not a science question but more like a geological background by the survey. The second and third aims are significantly different. Also, it's very important to highlight the research purposes and the novelties in the title, abstract and conclusion parts. Therefore, the detail demonstration of the connections between these two aims is highly expected. I actually suggest the authors to focus on one aim only in the paper.

The overall aim of this manuscript is to present a synoptic and holistic approach that we are convinced to be essential for the understanding of fluid dynamics, biogeochemical element cycling and ecology in the subsurface parts of the critical zone. To the best of our knowledge, this is the first paper that in deed presents such an approach in detail: It synoptically characterizes land use and land use history together with the soil and subsurface compartments (aeration zone, aquifers) involved in the flow of water and transport of matter in the subsurface. Moreover, the paper explains with two independent methods, how the subsurface is organized hydrostratigraphically, and how different groundwater compositions of distinct aquifer storeys are related to soils and surface land use in the groundwater catchment as well as to the geological structure, lithology/mineralogy and residence time of water in the subsurface. We rate this multi-method-approach as novel and important.
The guiding scientific question behind the paper is intrinsic in the aim of our manuscript and may read as follows: What are the essential and necessary information to be gathered in order to understand the factors that control dynamics and quality of a subsurface setting like the Hainich CZE?
Our holistic approach is novel as we, for the first time, describe in depth both aspects, the surface (recharge area reconstruction) and the subsurface (detailed aquifer stratigraphy). As such it is not a mere report on a hydrogeological survey (which we, indeed, frequently find in the literature). It is much more than this: A synoptic and detailed assessment of land use, pedology, geology and hydrogeology and its relations.
On top of this general aim we address four scientific questions.
(A) We describe the flow path functioning of a thin-bedded, mixed- carbonate/siliciclastic setting; this setting is widely distributed in marine successions (especially in the "Germanic Triassic" but also the carbonate rock formation in the Middle East i.e. Khuff formation) and hosts important groundwater resources. However, it was not adequately addressed in previous studies and described in literature, also due to lesser karstification, minor speleogenesis and moderate intrinsic vulnerability. To this end, typical karst studies mainly describe massive carbonates (i.e. the German Jurassic or Permian formation i.e. Zechstein carbonates) or exposed karst settings (i.e. alpine karst, exposed karst in SE Europe). We are sorry that we were not able to draw the reviewer´s interest on this important setting so far and we will explain the special characteristics and the importance of thin bedded carbonates.

(B) We present a new multi-method-approach for understanding hydrochemistry by characterizing the surface and subsurface. With the combined methods we are able to explain particular hydrogeochemical conditions within the multi-storey/hillslope aquifer system. For instance, the anoxic milieu in shallow aquifers and the oxic conditions in deep aquifers are closely related to their lithology/ out-crop patterns, penetrating karst features, land use and soil cover. We obviously did not strongly highlight this in the results chapter and will modify this.

(C) We surveyed all accessible subsurface compartments with a couple of tools/methods which were typically used for comprehensive vulnerability studies. In contrast to “anthropogenic hazard studies”, our focus lies on the reconstruction of the subsurface structure by utilizing groundwater quality data in addition to the lithological aquifer stratigraphy. In analogy with vulnerability studies, we determined the recharge areas and discussed groundwater quality in connection with the recharge area. We are convinced that our results are transferrable to vulnerability / anthropogenic hazard studies in comparable settings. We are sorry that we did not emphasize this in the motivation and the conclusions and will stress this with much more effort in a revised version.

(D) We demonstrate that a comprehensive investigation of aquifer connectivity in the transit/discharge area as well as soil cover and land use in the recharge area is necessary for understanding the groundwater quality and quantity in hillslope catchments. Our new approach encompasses (i) a detailed aquifer stratigraphy for understanding horizontal and vertical connectivity, based on scarcely available intact drill core data; (ii) a detailed geological investigation to understand that aquifer outcrop areas are definitely in the hillslope position and vertical infiltration is highly minimized by argillaceous confining beds (except in some sinkhole lineaments); (iii) a quantitative reconstruction of the aquifer recharge area: groundwater recharge takes place in a very small area; and finally (iv) a detailed investigation of soil properties in this recharge area. We are convinced that the understanding of groundwater quality and statistical analysis necessarily requires the knowledge of the abovementioned parameters in this type of thin-bedded carbonate/karst setting. Limiting the results to single aspects/aims, without presenting the multi-compartment survey data would have resulted in contradictions, because groundwater quality does not correlate with the land use directly surrounding the monitoring wells and groundwater physico-chemistry does not reflect the depths of the wells or the morphological positions (i.e. midslope, footslope); lastly vertical connections (horizontal/vertical flow) between different depth levels (aquifer storeys) cannot be understood by statistical analysis only; this requires a detailed drill core characterization.

As the reviewer got the impression of a sole hydrogeological survey report, we will revise the manuscript in order to countervail this impression.

Moreover, we decided to reorganize the manuscript such that the connection between the main aim and the three aspects, i.e.,

- Presentation of a new multi-method approach for understanding catchment area groundwater quality by reconstructing aquifer stratigraphy (and hydrogeochemistry) in the thin-bedded mixed, carbonate/siliciclastic setting of the Hainich CZE
- Assessment of recharge areas and surface-recharge properties (soil groups, land management) for determining intrinsic aquifer vulnerability and groundwater quality

- Characterization and interpretation of the vertical and lateral variability of karstification in a thin bedded carbonate-siliciclastic setting (intrastratal karst limited to single beds in some formations) becomes more obvious and intelligible also in the abstract and in the discussion sections. To further support this, we will emphasize more strongly the relationships between surface and subsurface influences on hydrochemistry in the discussion chapter. This includes to substantiate with our dataset the close relationship of controls of the groundwater quality by the recharge area characteristics.
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2) Because it is a research article instead of report, the authors are expected to explain why Hainich CZE is important and interesting to study.

We understand that the reference to the inaugural paper (Küsel et al. 2015) is not sufficient. We will introduce a section on the Hainich CZE which illustrates its importance and relevance, but also the uniqueness and capacities that are offered to the greater scientific community.

Are there any special geological characteristics? I'm not familiar to the hydrogeological setting in Germany, but I assume that carbonate-rock structures are widely distributed. Is Hainich CZE a typical karst aquifer in Germany? All of those are necessary to be fully illustrated in the manuscript.

The thin-bedded lithology and the hillslope tilting results in a confined-flow-setting with the following special properties: Main recharge takes place in outcrop areas. Because of the tectonic and exhumation history, the main aquifer recharge area is situated in the upper hillslope/summit areas. Thin soils only allowed forestry on hilltop. This resulted in favorable and low-impacted conditions, making the site interesting also for biodiversity studies. With respect to tectonics, the study site is relatively homogeneous and predictable, which allows us to track infiltrating precipitation from the recharge area to the groundwater in the aquifer.
We decided to present all important aspects of thin-bedded carbonate/siliciclastic settings hosting multi-storey aquifer systems, rather than compare this scarcely mentioned setting with well described prominent karst sites. Although, we applied methods, which are untypical for hydrogeology studies (geological mapping, recharge area reconstruction, soil mapping) and we detected large differences between aquifer storeys in directly overlying aquifer storeys, which wasn't discovered in old local site explorations. We agree that a general discrimination of thin bedded vs. "classical karst" settings is necessary for the reader. We will add a short comparison of general aspects and the consequences for aquifer quality, flow paths and intrinsic vulnerability to the discussion.

3) The authors used more than half of the words in this manuscript to introduce and describe the field works and data collections. Again, I would recommend the authors to focus on the discussion of statistical analysis (PCA and cluster analysis) of geochemistry data, and address the effect of karstification and hydrological stratigraphy on groundwater quality/hydrogeochemistry (section 4.2).

As we introduced a new and contrasting aquifer stratigraphy of a common, but barely addressed setting, we decided for a comprehensive method and results presentation. We are convinced that the multi-method approach is new to the understanding of groundwater systems. In our view, the description of surface properties, the subsurface structure and the groundwater chemistry results is necessary for understanding the connection between the surface signal and the aquifer quality. Nevertheless, we agree with the reviewer´s recommendation that a focused throng discussion of groundwater quality is also necessary. We will extend the discussion of the statistics with respect to surface (land use, soil) influences and subsurface influences. We agree that the relationship between karstification, aquifer stratigraphy and hydrochemistry is an important point, which will be added to the discussion chapter.

4) The authors mentioned the effects of fault zones on groundwater chemistry with dissolution-enlarged fractures.

The study area is characterized by old fault/fracture systems, which all have the same orientation (NW-SE) which is again parallel to the orientation of peripheral faults of low mountain ranges in Central Germany (Harz, Thüringer Wald, Hainich). These fault zones are described in regional literature as preferential flow paths, which is confirmed by a great number of karst springs (mentioned in the results chapter and piper plot), that are closely related to those fracture zones. Our descriptions of dissolution-enlarged fractures are limited to our drill cores material which had been recovered from wells which had not been drilled directly into fault zones. Nevertheless, we assume by analogy to our drill cores, that fractured/more permeable rocks are predisposed to a stronger karstification, resulting in a greater overall permeability in fault/fracture zones.

Hydraulic conductivity through the faults in karst aquifer can be larger in several magnitudes, due to the dissolution of carbonate-rock dissolution. Does dissolution play a more important role rather than faults? More explanations are expected.

As the described setting is an alternation of limestones and marlstones, dissolution is important for the bedding-parallel flow, although karstification effects are not very distinct (in comparison with classical karst/unconfined karst). Confinement by aquitard interbeds results in a strong limitation of karst to special formations (i.e. Trochitenkalk-formation) and to the lowermost intervals of these formations. At the base of this formation, karstification is very important for bedding-parallel flow. Other parts of the stratigraphic succession may be regarded more or less as fracture aquifers. By contrast, faults are important for the cross-formational flow, connecting aquifer storeys which are (apart from these localized zones) strictly separated. As we do not have drill cores from fault zones, we can only describe the relevance of dissolution and fracturing for the flow system. A direct comparison of permeabilities is not in the scope of our study.

We are aware that a distinction between fracture related and dissolution-related flow paths is important for the reader to understand the flow structure. We will describe the differences between these two preferential flow paths in the discussion chapter.

On the other hand, dual-permeability hydrological characteristics are commonly observed in karst aquifers.

As karstification and conduit-development is very limited in the described thin bedded carbonate-siliciclastic setting, as well as matrix porosity of dense marl and limestone rocks is assessed to very low (< 5 %), the aquifer system is not properly described with dual-porosity approaches of typical massive karst settings. We regret that our description of major flow paths lead to a misunderstanding. We will stress in the results chapter, that flow paths are predominantly fractures in the upper aquifer assemblage (fracture aquifers rather than karst cavities).

The authors should address some literature citations of flow properties in karst aquifer in the introduction.

This will be done accordingly

5) The authors might not have enough data, especially the historical data before the beginning of sampling. But it is interesting to see any trends of geochemistry data variation along time, with changes of land use type and anthropogenic factors.

The focus of this manuscript lies on the utilization of average hydrochemical data to support the aquifer stratigraphy. Both aquifer stratigraphy (based on lithology) and hydrochemical clustering (based on groundwater composition) are independent methods for demonstrating whether aquifer storeys are vertically separated or connected. In our view, hydrochemical fluctuations are not relevant to specific questions raised by this manuscript.
However, a 4-weekly sampled dataset of pressure heads and hydrochemistry is available for five hydrological years. As the pressure head and groundwater quality fluctuations are very complex (with respect to seasonality, rainstorm events and response times), the understanding of this system requires a lot of additional information (i.e. weather data, flow distances, permeabilities). For this reason we decided to discuss the dynamics (fluctuations) of the system in a second research article (in preparation), which will be based on the detailed flow structure/recharge area description of the present manuscript. We are aware that a brief outlook on the seasonality of groundwater quality is desirable for the reader to assess the mean values in the context of seasonal fluctuations. Therefore we could add an example chemograph (i.e. nitrate).

And a discussion of the effect of contamination/pollution/human factors to data is desired.

Hainich CZE represents a rare region in Central Europe that allows studying hydrogeochemistry in a non-contaminated, but used cultural landscape. Villages, waste disposal sites, industry and roads are not located within the groundwater recharge area. Although, specific pollutants are not measured, our hydrochemical analyses of major and minor cations/anions indicate a very extensive type of land management with very low concentrations in phosphate, ammonium, sulfur, potassium and heavy metals. As many of the major and minor ions are contained in soils, agricultural fertilizers, air pollutants and also in rocks (i.e. K, Na, S), a discrimination between natural and anthropogenic signals is very difficult. However we agree, that an extended discussion of “natural” and “anthropogenic” substances in the groundwater is essential for understanding the main message of this manuscript and will therefore be added to the discussion.

6) In the end of section 4.2, the authors classify three modes of subsurface water flow in the karst aquifer. I would say the lineaments of sinkholes are not necessarily due to flow through open faults.

Regarding the Upper Muschelkalk formations, karstification in thinly bedded limestones (as the most soluble material of this subgroup) is very limited and retarded, even though bedding-parallel flow maintain weathering and dissolution. The fracture networks and tight conduits do not cause sufficient mass deficits allowing hanging wall collapses. Even if deformed, or disrupted the stabilizing aquitard beds won’t be removed. The discussed sinkholes are “caprock sinkholes”, apparent in the caprock strata (Upper Muschelkalk) and caused by mass deficits by dissolution/subrosion in the underlying evaporite-rock formation (Middle Muschelkalk). Dissolution of evaporites in the underlying formation leads to passive collapses of the overburden limestone-marlstone strata (Upper Muschelkalk). Although the rock dissolution does not directly take place in the aquifer formation, this collapse structure certainly results in rock fracturing and an enhanced permeability in our target aquifers.

We are sorry that we did not define the type of sinkholes in detail and cause confusion to the reader. We will add a proper definition and explanation why we are convinced that cap rock sinkholes are relevant flow paths in the study area.

Is there a possibility that bedding parallel in either unconfined and confined aquifer can cause lineaments of sinkholes as well? Probably just track the faults/fracture zones from geological map/structure survey.

We agree with the reviewer, that caprock sinkholes may be related to bedding parallel flow which takes place in the evaporite formation below the carbonate-siliciclastic alternation that are the target aquifers in this study. We obviously failed to describe the nature of sinkholes in a convincing way and will modify this in the revision.
We mapped the surface phenomena (sinkholes and uvala-like depressions) in the field and on the basis of digital elevation models. Sinkholes are arranged in form of lineaments, which are parallel to the fault zones (connected to karst springs), described in regional literature. In our view, a good explanation for those caprock sinkholes is the penetration of surface water/shallow groundwater into

the underlying evaporite formation and the subsequent dissolution of evaporites.

7) Discussion 4.3 has weak relevant to the statistics analysis result. I don't think the authors have enough data to discuss karstification dissolution, so I recommend removing it.

As we have a very detailed dataset from karst phenomena in drill cores, we could show, that karstification takes place even in great distances to the surface recharge zones. This is relevant for the flow structure as all aquifer storeys are confined and vertical flow is to a great extent inhibited. The observation of karst phenomena in great distances to the recharge area therefor requires special processes providing CO_2 to the system. In our view, the discrepancy of intrastratal karst and the great flow distances falls in the field of this study, as we explain the surface influence on the aquifer/groundwater. We agree, that we did not properly describe the relationship between the surface-subsurface coupling theme and karstification. We transforme this chapter to a much shorter discussion of karstification-distribution subsequent to the hydrochemical discussion.

8) To be honest, I didn't get the key points in the conclusion part. The authors do not need to mention the results of mapping and survey in the conclusion part. I suggest the authors to summarize the results of data analysis and emphasize the relationships. It might be better to make the statements by bullet points.

Obviously we failed to explain the relationship between the interpreted results and the conclusions. A stronger emphasis on relationships between surface and subsurface is necessary. We will add to the conclusions, that our study shows, that groundwater quality in our setting / in shallow aquifers is to a large extent controlled by surface influx rather than by water-rock interactions. We will stress and summarize the relationship between land use, infiltration and hydrochemistry. We agree that bullet points are important for the reader, as the study aims and the flow structures in the study area are distinct compared to typical hydrogeological studies. For this reason we will use bullet points to structure the conclusions.