

1. RESPONSE TO ANONYMOUS REFEREE #1 (RC C2724)

Dear referee #1,

Thank you very much for the time spent reading our paper. We have carefully read the comments and remarks and revised the manuscript accordingly.

Here below, after each reviewer's comments, is a description (in black) of the new modifications made in the manuscript (modifications in the modified manuscript appear in red).

1.1.First Comment from referee #1

Review of "Time scales of regional circulation of saline fluids in continental aquifers (Armorican massif, Western France)" submitted by A. Armandine Les Landes et al. to Hydrology and Earth System Sciences (Hess). The first recommendation I would like to make is the very simple but very important addition to the title of this paper: insert the term "crystalline rock" after "continental", so that the title becomes "saline fluids in continental crystalline rock aquifers. I have been working a long time on saline fluids in continental sedimentary basins but have not closely followed the literature on waters in crystalline rock aquifers, such as the Armorican massif. If I had been paying more attention upon receiving the request for a review, I would have suggested the selection of someone else as a reviewer.

1.1.1. Response

We agree with your suggestion about the title of our paper and we inserted the term "crystalline rock".

1.1.2. Author's changes in manuscript

Thus on your advice the complete title will be:

"Time scales of regional circulation of saline fluids in continental crystalline rock aquifers (Armorican massif, Western France)".

1.2.Second Comment from referee #1

I would also include the term "crystalline rock" or crystalline basement" in the abstract and in key search terms.

1.2.1. Response

In the same way, we inserted these terms in the abstract and as a keyword (see below).

1.2.2. Changes in manuscript

Lines 30, 37 and 43

Abstract

In recent decades, saline fluids have been sampled worldwide at great depths in continental basements. Although some of them have been attributed to marine transgressions the mechanisms allowing their circulation is not understood. In this paper, we describe the horizontal and vertical distribution of moderately saline fluids (60 to 1400mg/l) sampled at depths ranging from 41m to 200m in **crystalline rock** aquifers at the regional scale of the Armorican Massif (northwestern France). The horizontal and vertical distributions of high chloride concentrations are in good agreement with both the altitudinal and vertical limits and succession of the three major transgressions between the Mio-Pliocene and Pleistocene ages. The mean chloride concentration for each transgression area is exponentially related to the time spanned until present. It defines the potential laws of leaching of marine waters by fresh meteoric waters. The results of the Armorican aquifers provide the first observed constraints for the time scales of seawater circulation in the continental **crystalline** basement and the subsequent leaching by fresh meteoric waters. The general trend of increasing chloride concentration with depth and the time frame for the flushing process provide useful information to develop conceptual models of the paleo-functioning of Armorican aquifers.

Keywords: Saline fluids, **crystalline** basement, groundwaters, paleohydrogeology, geologic time scale, climatic events.

1.3.Third comment from referee #1

I have read through the manuscript, and the research results seem both reasonable and well-presented. However, it would have been useful to have included some of the isotopic results mentioned in passing on p. 6607, line 18, particularly oxygen-hydrogen systematics.

1.3.1. Response

I agree that it would have been useful to include some of the isotopic results. Nevertheless, the addition of isotopic results in this paper could provide more complexity that does not seem necessary. These isotopic results are presented and described in another paper: “Impact of climate changes during the last 5 million years on groundwaters in basement aquifers“ recently submitted by Luc Aquilina (co-first author of this paper) to PNAS (Physical Sciences / Earth, Atmosphere and Planetary sciences).

I therefore suggest adding the previous reference where the readers will find a detailed description of isotopic results, analytical methods, interpretation of isotopic measurements and complementary information on the paleohydrogeology of the Armorican aquifers.

Moreover, according to your following comment, you will find complementary information on mechanisms that explain the displacement and the mixing between meteoric and marine waters after a transgression (see response below).

1.3.2. Changes in manuscript

The reference Aquilina et al. 2014 has been added. Line 241

1.4. Fourth comment from referee #1

I can see influx of marine waters into the system by density-driven flow during marine transgressions. It is less clear to me from the manuscript how marine waters become displaced or how mixing occurs with meteoric waters after a transgression.

1.4.1. Response

As stated above, complementary information can be found in the paper: “Impact of climate changes during the last 5 million years on groundwaters in basement aquifers“. However, the displacement of marine waters and the mixing occurring with meteoric waters are the underlying processes used in the two conceptual models to explain the paleo-functioning of Armorican aquifers.

During marine transgression, seawater was introduced into the basement by density-driven flow. This mechanism induced the displacement of former fresh groundwater by seawater. Then due to diffusion process taking place on million year (according to the time since transgression) the diffusion length scale (few hundred meters) leads to a perfect mixing between marine waters and freshwaters already present in the whole rock porosity. Third, following transgression, the marine signature contained in the system is flushed out by groundwater circulations fed by meteoric waters. The preservation of marine signature throughout the Armorican basement and the clear increase with depth underlines the limited downward fresh groundwater circulation loops. But, the flushing process leads to the displacement of marine waters in the upper part of Armorican aquifers. The limited depth of groundwater circulations is explained by the dense hydrological network, the high sea-level and the low topography relief within the Armorican massif and characterizes the salinity increase with depth.

1.4.2. Changes in manuscript

Sentences have been added Line 413-425

2. RESPONSE TO ANONYMOUS REFEREE #2 (RC C4938)

Dear referee #2,

Thank you very much for the time spent reading our paper. We believe that these remarks contributed to improve the manuscript.

2.1. First comment from referee #2

More explanation is needed to justify and explain the value of C_{in} parameter. It seems that was determined from the fitting of the equation to the empirical data (3 points). If that is correct, the “fitted” C_{in} value seems poorly constrained. What range of values might a sensitivity analysis have determined to be likely? Could a probability distribution be roughly assigned to C_{in} value. Do the authors have an a priori reason to believe that C_{in} should be close to 100 mg/L?

2.1.1. Response

Indeed, the value of C_{in} parameter was determined from the fitting equation to the empirical data. The fitted curve (simple exponential function) is constrained by 3 parameters: the current chloride concentration of the submerged area (c), the age of the transgression that has flooded the area (t_e) and the background concentration (c_0) which corresponds to the chloride concentration of area not influenced by past marine transgression. Explanation has been added to justify the range of parameter values.

2.1.2. Changes in manuscript

Line 312-316

2.2. Second comment from referee #2

An interesting implication of the reasoning is what would have been the Cl concentration in the aquifer at the end of the transgression. I believe there is little in the literature about this question; did the authors’ literature review find anything regarding seawater mass emplacement? When there has been a major marine transgression lasting XX millions of years, would the subsea aquifer have been flooded by seawater? If so, would not the C_{in} value be Cl= 19,000 mg/L? The authors should expand their reasoning for why the initial post-transgression aquifer Cl would have been two orders of magnitude less than seawater salinity. Do they have a quantitative explanation for why emplacement of seawater under prolonged transgression would have been incomplete?

2.2.1. Response

During transgression, seawater is introduced into the basement by a simple mechanism driven by density and gravity process. This mechanism drives the displacement of former fresh

groundwater by seawater, but this process enables the saline fluid circulation at great depth in fractured aquifers through the most permeable structures. Nevertheless, all scales of porosity containing former fresh water are not directly influenced. The presence of saline fluid in all scales of porosity requires the diffusion of saline fluid on long term. Thus, the former fresh water already presents has a great contribution on the injected concentration value. Considering the presence of fresh water in the porosity of aquifers a concentration of 19000 mg/L for the aquifer flooded by seawater is clearly overestimated. Moreover, the C_{in} value estimated in our study is related to the entire volume of the aquifer which tends to reduce the value. To finish, as mentioned in the manuscript, if the increase of chloride concentration with depth is a systematic evolution (which appears to be the case as demonstrated by the general trend), a deeper sampling within Armorican aquifers could then give higher C_{in} values.

2.2.2. Changes in manuscript

Sentences have been added Line 376-384

2.3. Third comment from referee #2

The authors mostly call tau (τ) "residence time." I am not sure it is residence time (or storage time), as usually defined [(total mass of solute in basin)/mass flux]. tau (τ) seems more like a half-life, a rate constant, but not exactly analogous. This meaning needs to be more carefully and thoroughly considered. It is the crux and most interesting part of this paper! Implications: how might tau (τ) differ between basins? What physical parameters and evolution of paleohydrologic boundary conditions might affect the distribution of tau (τ) among aquifers?

2.3.1. Response

The term "residence time" is usually used in the context of the origin of salinity investigations. One of the main examples is the study: Origin and residence time of salinity in the Äspö groundwater system, where the residence time of the deep salinity is estimated from the long half-life of ^{36}Cl radionuclide. Even if this term strictly corresponds to a half-life, the residence time term is used as analogous in the literature.

However, I agree that the most interesting part of the paper is the residence time frame. A discussion about the variations of τ between basins is necessary. Sentences have been added about the implications of physical parameters and paleohydrologic conditions that may have influenced the residence time value.

2.3.2. Changes in manuscript

Line 389-407

2.4.Fourth comment from referee #2

Three field-based data points makes for a weak fit of an exponential equation. In my comment 1 above I hint that additional discussion on uncertainty in the fitted equation is needed and appropriate. But there is another source of information that the authors could use, which comes from analytical (easily accessed or redone) and numerical models (no so available). For example, Domenico and Robbins (1985. The displacement of water from connate aquifers. GSA Bulletin 96:328) and surely others since define analytical solutions for a similar problem. The authors could adapt such an approach, and then "sample" the domain through time and generate many more than three data points. Wouldn't that comparison be worthwhile (and relatively easy)?

2.4.1. Response

I agree that the three field-based data points makes for a weak fit. However, it is important to note that situation where three areas have been submerged successively by marine transgressions is exceptional. Even if, the fit is based on three points the contribution provides on the saline fluids residence time is clearly better than all previous studies and consequently constitutes a strong constraint.

Another option is to use another source of information and currently represents the next step of this study. As mentioned (line 452) the next step of this study will to test the functioning hypothesis with numerical modeling in order to provide more realistic models. The ongoing numerical modeling work will provide to generate more than three points. Thus, as you mentioned the comparison will be useful but in my view constitutes a complementary approach that requires a study mainly focusing on numerical models. In this way, analytical solutions may be used for the comparison of numerical modeling and field data.

2.4.2. Changes in manuscript

Sentences have been added. Line 369-373

2.5.Other comments from referee #2

Comments 1

1. Is leaching of marine waters best term? Displacement? Does leaching have specific content not relevant to this hydrologic context?

Response 1

I agree that we could use another terms to qualify this hydrologic context. Indeed, from the literature, it appears that there is no specific term to explain this hydrologic context. The terms displacement or leaching or flushing of marine waters are used to describe the replacement of paleoseawater by fresh groundwater.

Changes in manuscript

The term “displacement” has been added in parentheses in the abstract after the leaching term to find all different terms (Flushing/leaching/displacement) at the beginning.

Comments 2

2. P. 6601 versus p. 6605. Is there any reason to assume that there were no transgressions older than Mio-Pliocene that could have emplaced seawater in these ancient rocks. No sedimentary record of older transgressions. Or only the most recent ones count (p. 6605). Might be worth addressing more clearly.

Response 2

On the contrary, there are sedimentary records which justify there were transgressions older than Mio-Pliocene. But, as explained only the most recent ones is important in our context. To clarify this concept, a sentence has been added earlier in the paper to provide details on the objective of the methodology.

Changes in manuscript

Line 74-80

Comments 3

3. P. 6607 (6.1). Both ‘salinities’ (60 to 1400 mg/L) and ‘chloride concentration’ are used but it is not clear that the authors are not treating them as synonymous. Is the meaning of salinity = total dissolved solids as used?

Response 3

To avoid ambiguity between “salinity” and “chloride concentrations”, a sentence has been added to specify that both terms salinity and chloride concentration are treating as a synonymous (Line 240). Chloride, is one of the major inorganic anions in saltwater. Although salinity is a measure of the total salt concentration, including principally Na and Cl ions, in the context of our study as chloride is clearly of marine origin salinity is used as analogous.

Comments 4

4. P. 6608. There are many other sources of chloride than the 3 listed. The 3 might be the only ones relevant to this study, but the others should be recognized. The most obvious are solution of halite (there are no evaporates in the basins?) and evaporatively concentrated brines or other formational brines at depth.

Response 4

We agree that many other sources of chloride exist (as evaporitic deposits of geological formation...). But in our case, the relationship Br/Cl shows that plotted versus chloride, bromide contents fit with the seawater dilution straight line which explain the marine origin. While the relationships between chloride and bromide will be completely different for evaporated brines or evaporite leaching.

Changes in manuscript

Sentences have been added to clarify this point. Line 265-268.

Comments 5

5. P. 6612 "All previous studies" is a vague reference. Is that all as in ALL, including the whole literature cited and not cited in this paper? Or does it refer to just the papers cited in this paper? Could be made explicit by reciting the relevant papers.

Response 5

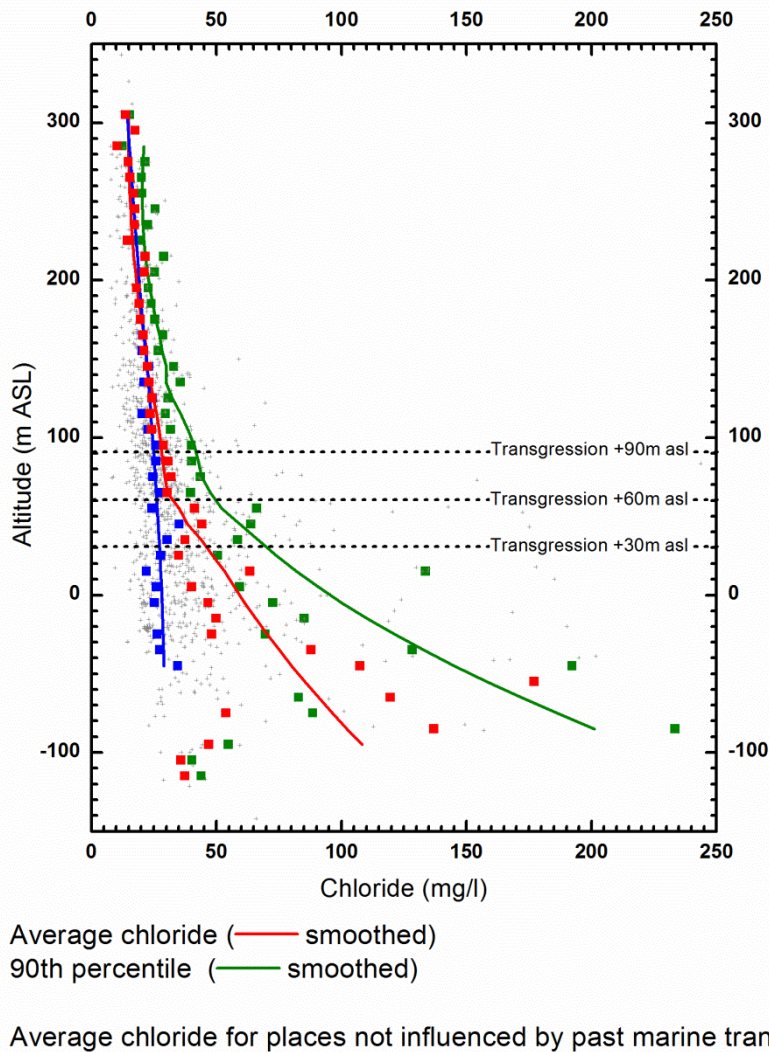
To be more specific, the references have been added after "All previous studies".

Comments 6

6. Figure 6. I don't believe the first derivative adds anything to the paper and that part of the figure should be deleted.

Response 6

We agree that the first derivative is not primordial. Thus this part has been deleted from the Figure 6. The sentence associated has been deleted. And caption has been updated.



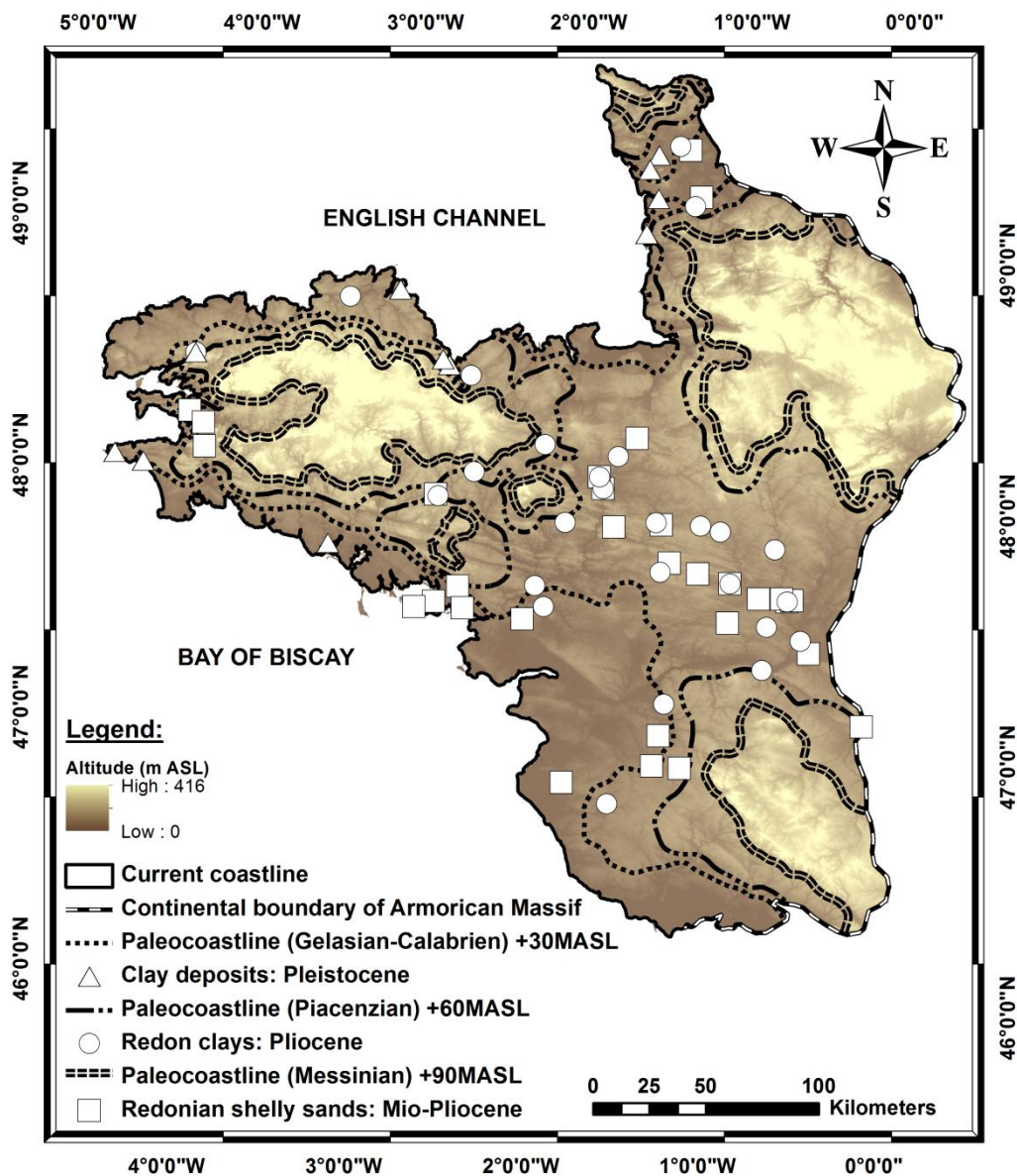
Comments 7

7. Figure 3. Is there something mislabeled? Caption cites 'characteristic sediments' but the explanation and map do not appear to indicate sediment information. Do the three symbols (triangle circle square) represent age of sediment? If so, caption should state 'age of sediment' and not 'characteristic sediments.' The latter would suggest shale, sandstone, etc.

Response 7

Figure 3 has been modified (characteristics sediments for each transgression have been indicated).

Changes in manuscript



Comments 8

8. Figure 5. Because there is a 3D distribution of chloride concentration, I am not sure the value of displaying this information on a 2D maps. I suggest deleting this figure; coastlines are in figure 3. Authors should explain to the editor justification for keeping figure 5.

Response 8

I do not agree with the deletion of the figure 5 for two main reasons. (1) This map shows clearly the correlation between the spatial distribution of high chloride concentrations and the area submerged by each transgression. (2) This figure is important because it provides the basis to establish the exponential fit and will facilitate understanding of all readers.