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Time scales of regional circulation of saline fluids

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

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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 1400 mg L⁻¹) sampled at depths ranging from 41 to 200 m in 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 sea-water circulation in the continental 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.

1 Introduction

In recent decades, saline fluids have been sampled at great depths (0.5–5 km) in continental basements (Bucher and Stober, 2010; Frape et al., 2003). In several places, these fluids have been considered as old seawater introduced in aquifers during marine transgressions (Aquilina and Dreuzy, 2011; Beaucaire et al., 1999; Bottomley et al., 1994, 1999; Douglas et al., 2000; Greene et al., 2008; Louvat et al., 1999; Négrel and Casanova, 2005). Two alternative origins related either to glacial recharge of brines due to cryogenic mechanisms (Starinsky and Katz, 2003), or to anthropogenic sources (Kelly et al., 2008; Mullaney et al., 2009; Panno et al., 2006; Perera et al., 2013) have

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also been proposed. These three hypotheses have led to extremely different estimates of the saline fluid residence time (Devonian or Pleistocene for the Canadian brines, for example) and thus of the paleohydrogeology of the continental aquifers. Although thorough investigations have been carried out in several places, the origin and fate of the salinity are still issues. We have almost no field observations identifying the potential mechanisms leading to marine fluid introduction and conservation in hard rock aquifers, as well as the influence of glacial recharge at various depths in these aquifers remain. Furthermore, the time-scales of these mechanisms are unknown.

In this study, we investigated the effects of marine transgressions in the spatial (both horizontal and vertical) distribution of chloride concentrations at a regional scale. Since the transgression characteristics differ from one event to another, we expect to find higher concentrations in the areas flooded by the later. The decrease of chloride concentrations with time potentially provides constraints for fluid circulation time-scales in the basement.

The study focuses on the Armorican basement (western France), where several authors have reported evidence of saline fluids (Ayraud et al., 2008; Pauwels et al., 2010). Three main transgressions were recorded during Mio-Pliocene to Pleistocene times, the latest being the highest. This is a particularly favorable context since a large part of the area flooded by the oldest and highest transgression remains unaffected by later transgression events.

2 Geological background

The Armorican basement extends over an area of 68 500 km² in the northwestern part of France. The Armorican massif is a crystalline basement localized between the English Channel and the Bay of Biscay. The northern part corresponds to the Cadomian orogenic belt (Chantraine et al., 2001). The central and southern zones belong to the European Variscan belt (Le Corre et al., 1991). The Armorican massif is a basement made of Upper Proterozoic to Paleozoic formations mainly composed of low and

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layer 20 to 30 m thick is present in many places, and likely ubiquitously (Brault, 2002; Dewandel et al., 2006). Roques (2013) has studied the influence of fault zones for sites characterized by high groundwater yields at the scale of the Armorican massif. At this scale, groundwater resources in the shallower part (<20 m) are widespread but limited in term of productivity. Conversely, high groundwater productivities are associated to fault zones below the weathered zone. Indeed, groundwater resources at significant depth have been found in the Armorican bedrock (Le Borgne et al., 2006; Leray et al., 2013; Roques et al., 2014) and it can be considered that the fractured system constitutes a water reservoir in the continental basement. Free groundwater circulation in crystalline environments has been reported from several studies (Banks et al., 1996; Larsson, 1987). Several studies have shown that basement is generally characterized by an interconnected fracture system at several km depth (Aquilina and Dreuzy, 2011; Edmunds and Savage, 1991; Stober et al., 2002). Hydrodynamic properties of crystalline rock aquifers of the Armorican basement have been intensively investigated in several research sites (Ploemeur and St-Brice-en-Cogles sites, Fig. 2) (Le Borgne et al., 2004, 2006; Dorn et al., 2012; Leray et al., 2013; Roques, 2013; Roques et al., 2014; Touchard, 1999) (Fig. 2). Moreover, previous studies in the Armorican basement have identified a compartmentalization of these crystalline rock aquifers from hydrological, geochemical, geological and geophysical data (Ayraud et al., 2008; Roques, 2013; Roques et al., 2014), and from regional-scale numerical modeling (Goderniaux et al., 2013). Geochemical studies also showed specific chemical compositions of the water in the deep fractured aquifer, potentially including saline groundwaters (Aquilina et al., 2013; Ayraud et al., 2008; Pauwels et al., 2013).

4 Chloride database

The quantity supplied and the quality of the main groundwater resources in the Armorican massif are monitored through a public well network (ADES: “banque nationale d’Accès aux Données sur les Eaux Souterraines”, National database for groundwater

Pliocene: Reuverian ($\sim 2.7 \pm 0.3$ Myr) with sea level +60 m a.s.l.; and the most recent is Pleistocene: Gelasian/Calabrian ($\sim 1.8 \pm 0.2$ Myr) with sea level +30 m a.s.l.

Using local studies on the large-scale relief development and the paleotopographic evolution of the Armorican basement (Bonnet et al., 1998, 2000; Brault et al., 2004; Guillocheau et al., 2003; Lague et al., 2000; Lenôtre et al., 1999; Morzadec-Kerfourn, 1997; Van Vliet-Lanoe et al., 1998), a paleotopography was reconstructed for the whole area.

Globally, the topography evolution is the result of tectonic movements, climate and eustatic variations since the Mio-Pliocene. The modern topography is characterized by of incised valleys with an incision average depth of 60–100 m and significant topographic variations. The onset of the current incised and dense hydrological network is probably not older than 1 Ma (Bonnet et al., 1998, 2000; Lefebvre et al., 1994). This topography contrasts with the smooth planation surface of the Mio-Pliocene (Brault et al., 2004). The development of the Pleistocene topography of the Armorican massif is explained by vertical movements due to the N160 compression generated by the Alpine collision (Bonnet et al., 1998; Müller et al., 1992; Nicolas et al., 1990). This differential tectonic uplift has been estimated from geomorphological studies (Bonnet et al., 1998, 2000; Lague et al., 2000). Incision is measured using a digital elevation model analysis. A distributed field of incised quantities is produced for each individual drainage basin (Jost, 2005). Moreover, relief induced by fluvial incision partly reflects continental movements and can be used to estimate uplift. Indeed, considering that the drainage networks adjust to sea level, the induced incision has the same magnitude in different basins. Thus the difference of incision is used to estimate the relative amounts of differential uplift between basins. The estimation of differential uplift for each basin has been grouped by zone due to the regional organization. A differential tectonic uplift has been observed for the western part compared to the eastern part, along the Quessoy/Nort-sur-Erdre fault (Brault et al., 2004; Jost, 2005).

From these previous work, to reconstruct the paleotopography of the whole area, we first used a digital elevation model (DEM, 100 m resolution) given by the Institut

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chloride concentration for each area is inversely related to the time elapsed since the last corresponding marine transgression.

This relationship shown in Fig. 7 can be fitted by a simple exponential function of the general form:

$$c = c_{\text{in}} \exp\left(-\frac{t_e}{\tau}\right) + c_o, \quad (1)$$

where c is the current chloride concentration, t_e is the age of the last transgression event, τ a characteristic time scale, c_o a background concentration, and c_{in} an additional concentration injected at the time t . Equation (1) is the solution of the differential equation:

$$\frac{dc}{dt} = -\frac{(c - c_o)}{\tau} + c_{\text{in}}\delta(t_e), \quad (2)$$

$\delta(t)$, the delta function, simulates quasi-instantaneous injection of chlorides at transgression times. The background chloride concentration c_o can be measured in places not influenced by past marine transgression; it is estimated to $21.5 \pm 0.3 \text{ mg L}^{-1}$. The regression fit leads to a time constant τ of $2.3 \pm 0.1 \text{ Myr}$, and an injected concentration of $100 \pm 14 \text{ mg L}^{-1}$. This residence-time order of magnitude is consistent with the residence time of several million years suggested by (Thury et al., 1994) for deep water as well as several studies supporting long residence times required by “fossil” seawater (Stober and Bucher, 1999). It can be noticed that these studies of fluids containing a paleoseawater end-member have chloride concentrations with an order of magnitude similar to those presented here.

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7 Discussion

7.1 General trend of increasing salinities with depth

The main results of this paper present clear evidences on the origin and the age of saline fluids in the continental crust at the scale of the Armorican massif. We report the vertical distribution of chloride concentration which presents a concentration increase with depth at the Armorican massif scale. Such a trend has already been observed in several places throughout the world in continental crust.

Highly saline brines have been found in various geological environments in the Canadian Shield (Frape et al., 1984; Fritz and Frape, 1982; Gascoyne and Kamineni, 1994), in the crystalline basement of Europe (Black Forest in Germany, Soultz-sous-forêt in France) (Aquilina et al., 1997; Pauwels et al., 1993; Stober and Bucher, 1999), in the Fennoscandian shield (Fritz, 1997; Nordstrom et al., 1989) and in the England Carnmenellis Granite (Edmunds et al., 1985). Saline fluids sampled in these sites report a wide range of chloride concentrations. At the surface, chloride concentration ranges from around ten to hundred mg L^{-1} , at much greater depth around 500 m concentration values are between one hundred to thousands mg L^{-1} and below 1 km depth concentrations range from hundreds to tens of thousands mg L^{-1} .

For all these sites chloride concentration have been analyzed by intervals of depth where the average chloride concentration is calculated in order to define the concentration gradient until approximately 1 km of depth (Fig. 8). The Armorican data are also plotted on this graph (Fig. 8). All the studies show a general increase of salinity with depth. In the first three hundred meters a high gradient of salinity is observed and between 350 and about 700 m, another gradient is observed. Below this depth, a stabilization seems to be observed although there are relatively few data (Fig. 8). However the main difference between all studies relies in chloride concentrations of groundwater samples at shallow depths. In fact, chloride concentration can vary by one order of magnitude, with concentrations around ten to hundred mg L^{-1} , as for the Armorican basement and the Canadian Shields, respectively (Fig. 8).

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that chloride flushing from the aquifers is a process requiring approximately 2.3 Myr to return to initial conditions. Despite uncertainties on paleocoastlines and the exponential function arbitrarily chosen, the essential point is the clear evidence of past marine transgression during the last millions years and thus leaching processes being active during a similar duration. We use these characteristic time of hydrodynamic processes to constraint the conceptualization of basement aquifers functioning in the two next sections.

7.3 Conceptualization of Armorican aquifers paleohydrogeology

The Armorican massif presents clear evidences of a marine origin of the saline component in the fluids at the massif scale. The general trend of increasing chloride concentration with depth supports the seawater introduction by gravity-driven flow at depth in the basement during past marine transgressions followed by a flushing phase with freshwater. Moreover, the chloride releasing rate provides a time frame for the flushing process which has been estimated in the order (of magnitude) of a few million years. Such information can be used to develop conceptual models of the paleo-functioning of Armorican aquifers. The objective of this section is thus to propose conceptual models that may account for the chloride increase with depth on one hand and for the chloride concentration increase fit with time presented above.

The crystalline basement of the Armorican massif presents aquifers with a high transmissivity associated to fault zones (Roques et al., 2014). Considering the current topography of the Armorican massif including three domains of higher elevation above 200 m, the groundwater flow system is driven gravitationally which leads to both local and regional circulation loops (Toth, 1963). Considering the modern situation and constraints provided by the distribution of chloride discussed previously, two main possibilities may explain the distribution of saline fluids in Armorican aquifers.

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7.3.1 First conceptual model: perfectly mixed aquifer

First, the crystalline rock aquifers are considered as a perfectly mixed reactor where the time constant of millions of years is also the residence time of chlorides in the system. The chloride contained in the system is slowly flushed out by freshwater under the influence of topography-driven groundwater flow (Toth, 1963). This model requires that chlorides are located in a compartment of the groundwater system whose recharge is far lower than the current recharge rate (around 200 mm yr^{-1}) at the top boundary of the system (see Appendix A). This does not necessarily mean that this conceptual model is not consistent with data. On the contrary, it seems consistent with the flow partitioning between local and regional flow systems initially presented (Toth, 1963) and precised more recently (Cardenas, 2007; Goderniaux et al., 2013). The characteristics of the partitioning between shallow aquifers and deep aquifers have been studied using the topography of Brittany and assuming a constant permeability with depth (Goderniaux et al., 2013). This study shows four interesting results about the partitioning between shallow (short circulations related to first-order basins) and deep (more regional circulations) aquifers: (i) the “deep” compartment can be quite shallow depending on local topography characteristics; (ii) the residence time of particles is well fitted by an exponential function, which means that the concentration flushing should behave similarly; (iii) the partitioning depends on the total recharge; (iv) the recharge flow in the deep aquifer is 2–5 times smaller than the total recharge. These recharge values are not small enough to explain the 2.3 Myr residence time observed for chlorides, which could mean either that the first model is inconsistent with data, or that the assumption of Goderniaux et al. (2013) is unrealistic. A permeability decrease with depth, may be observed in the continental crust with a likely factor of 100 within the first kilometer (Ingebritsen and Manning, 1999; Saar and Manga, 2004; Stober and Bucher, 2007). Such decrease could account for a much smaller recharge of the deep groundwater system and thereby a flushing process less efficient. Through modeling is ongoing to further test this hypothesis.

7.3.2 Second conceptual model: deep reservoir

In the second case, the presumption is that chlorides could be localized in a deep reservoir i.e. in the deeper part of the aquifer. Then chlorides would be leached out from the aquifer system after diffusion from the deep reservoir towards the active part of the crystalline rock aquifers where leaching is efficient. Calculations for this conceptual model (see Appendix 8), lead to a “salt” layer thickness of 40 m that does not seem unrealistic as regards the chloride evolution with depth observed. However this conceptual model would predict very high concentrations at depth that are not yet observed in the Armorican basement in the absence of very deep drillings. But, this could be in agreement with highly saline fluids sampled in various geological environments at great depths worldwide (Fig. 8). As in the crystalline basement of the Black Forest where Stober and Bucher (1999) suggest that the deep salt water discharges is related to the upwelling of deep water which may bring up chloride from a deep reservoir. However, it can certainly be affirmed that the salt contained in the deepest part is not solid due to the chloride to bromide relationships which excludes this possibility.

The next step of this study will be to test these two functioning hypothesis with numerical modeling in order to provide more realistic models. The objective is to provide some constraints on the saline fluid circulation at the regional scale as well as to understand if the heterogeneity and hydrodynamic conditions may induce a wide range of leaching rates which could explain the large range of chloride concentrations observed.

8 Conclusions

Chloride concentrations in groundwater were investigated in the Armorican basement at shallow depth (from +350 to -150 m a.s.l.). The high concentrations observed in some places (from 60 to 1400 mg L⁻¹) are shown to originate from marine transgressions. The distributions of high chloride concentrations are in good agreement with both the altitudinal and vertical limits and succession of the three major transgressions

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between the Mio-Pliocene and Pleistocene ages. During each of the last transgressions, seawater was introduced by gravity-driven flow at great depth in the basement. This seawater end-member has only partially been flushed by meteoric waters (over 1 to 2 million yr) between successive marine transgressions, and since the last one. Considering the paleocoastlines, three zones have been affected by the transgressions. The average chloride concentration calculated for each area is inversely related to the time elapsed since last marine transgression. This relationship between concentration and transgression age can be fitted by a decreasing exponential function which indicates that chloride flushing from the aquifers is a process requiring approximately 2 Myr. The fate of chloride is conceptualized using two main models: a dilution or leaching of a deep reservoir. Both models provide information on the history of the saline fluids in fractured aquifers. The first model requires a drastic partitioning between shallow and deep aquifers. In the second model, chlorides are supposed to be released by diffusion from a deep reservoir, leached by fresh groundwater flow and bringing up to the shallower aquifer. The model supposes a deep reservoir, which has not yet been identified in the Armorican massif, but could exist. This study thus provides important constraints on the seawater circulation in hardrock aquifers at a continental-scale area.

Appendix A:

The objective of this section is to develop simple calculations that aim at figuring out the meaning of the data and fit presented before. We first assume that the average of chloride concentrations measured is related to the average chloride concentration in the aquifer used in the model. Because of the increase of chloride concentration with depth and because of the limited sampling depth, it is unreasonable to pretend that the average of the measurements equals the chloride average concentration in the entire aquifer. Thus, we just guess that the chloride average in the entire aquifer evolves with a similar time scale to the measured averages, which occur for instance if the depth dependency function is similar in different places. Regardless of the model, the general

evolution of the chloride concentration is given by a mass balance equation of the type:

$$\frac{d(V\bar{c})}{dt} = -Q_c + Q_{in} \quad (\text{A1})$$

where V is the system volume, \bar{c} the average chloride concentration in the system, Q_c the total flow at the system discharge boundary, and Q_{in} the total flow at the system recharge boundary. The “system” can be the entire aquifer or any part of it. Q_{in} represents a potential chloride inflow from the recharge boundary. If we assume a background concentration c_o , Q_{in} can be written as:

$$Q_{in} = \iint_{S_r} r(s) \cdot c_o ds = c_o \cdot A_r \cdot \bar{r} \quad (\text{A2})$$

where S_r is the system recharge boundary, $r(s)$ the recharge at any point s of the boundary, A_r the recharge boundary surface, and \bar{r} the average recharge flow.

We then define two end-member models that may explain the chloride evolution in basement aquifers.

A1 The perfectly mixed aquifer

In the first model, the chloride contained in the deeper part of the aquifer is slowly flushed out from the system along the discharge at a rate Q_c such as:

$$Q_c = \iint_{S_d} q(s) \cdot c(s) \cdot ds \quad (\text{A3})$$

where S_d is the system discharge boundary, $q(s)$ the flow discharge at any point s of the boundary, and $c(s)$ the chloride concentration. In a perfectly-mixed reactor, the total concentration at the boundary is equal to the concentrations average. Even if this approximation is certainly unlikely in highly heterogeneous aquifers, we assume that

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h' , which is a reasonable assumption if the aquifer thickness is small compared to its horizontal extent Q_c is defined as :

$$Q_c = O' \cdot A \cdot D \cdot \frac{(\bar{c} - c_o)}{h'} \quad (\text{A6})$$

with O' an “order-of-magnitude” constant similar to O . The influx Q_{in} is now nil and the general equation writes as:

$$\frac{d\bar{c}}{dt} = \frac{(c_o - \bar{c})}{\tau'} \quad (\text{A7})$$

with $\tau' = O'^{-1} \cdot \frac{h'^2}{D}$.

The molecular diffusion D_m of chloride in water is of the order of $2 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$ (Li and Gregory, 1974; Wang et al., 1953). Considering a porosity of 1 %, the diffusion D of chloride is of the order of $2 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$. A time constant of 2.3 Myr is thus consistent with a salt layer thickness of about 40 m.

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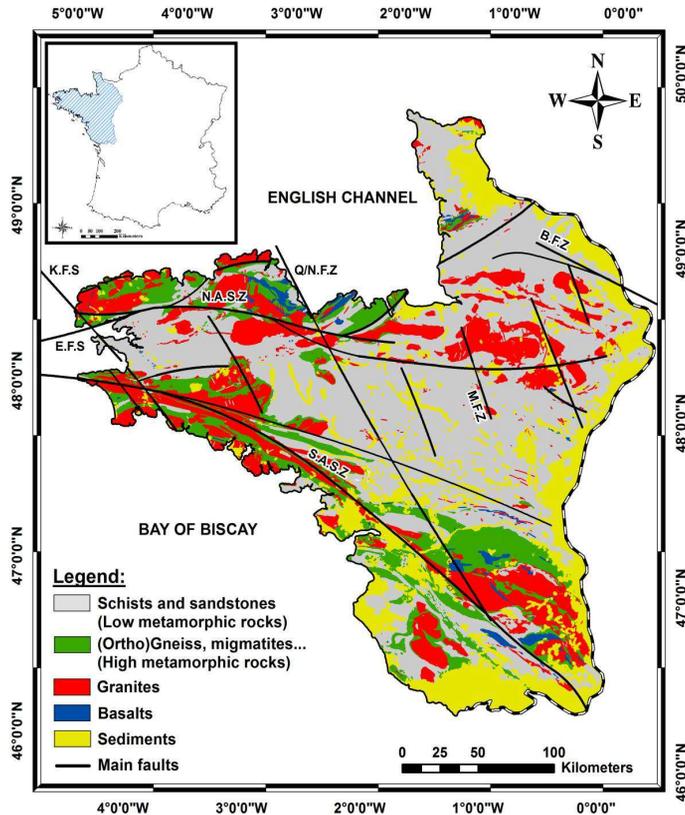


Figure 1. Simplified geological map of the main lithological units and main geological structures of the Armorican Massif: NASZ: North American Shear Zone; SASZ: South Armorican Shear Zone; Q/NFZ: Quessoy/Nort-sur-Erdre Fault Zone; MFS: Mayenne fault System; KFS: Kerforne Fault System and EFS: Elorne Fault system (according to Bonnet et al., 2000).

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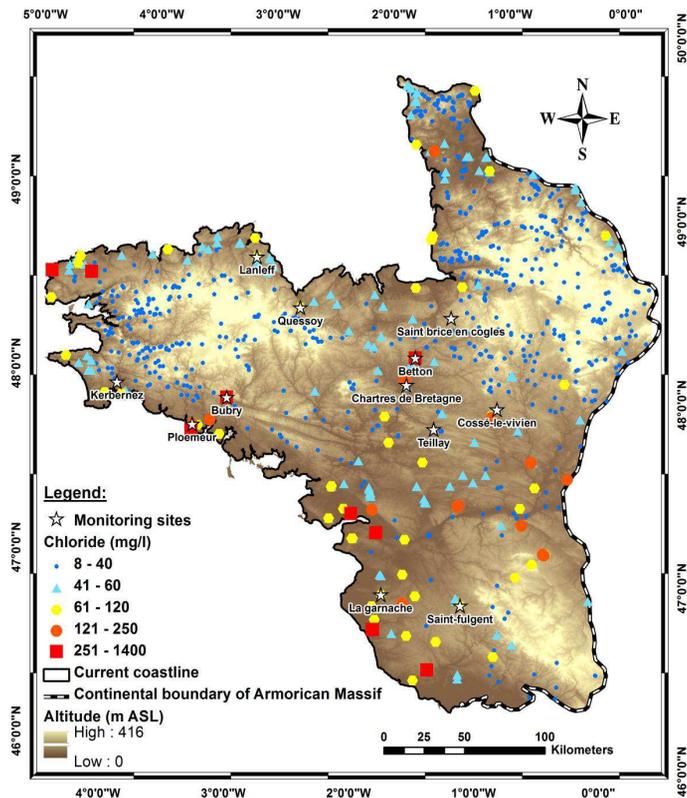


Figure 2. Map of the Armorican Massif including the distribution of chloride concentration for the whole area (from preprocessed chloride database) and the location of the 12 sites investigated.

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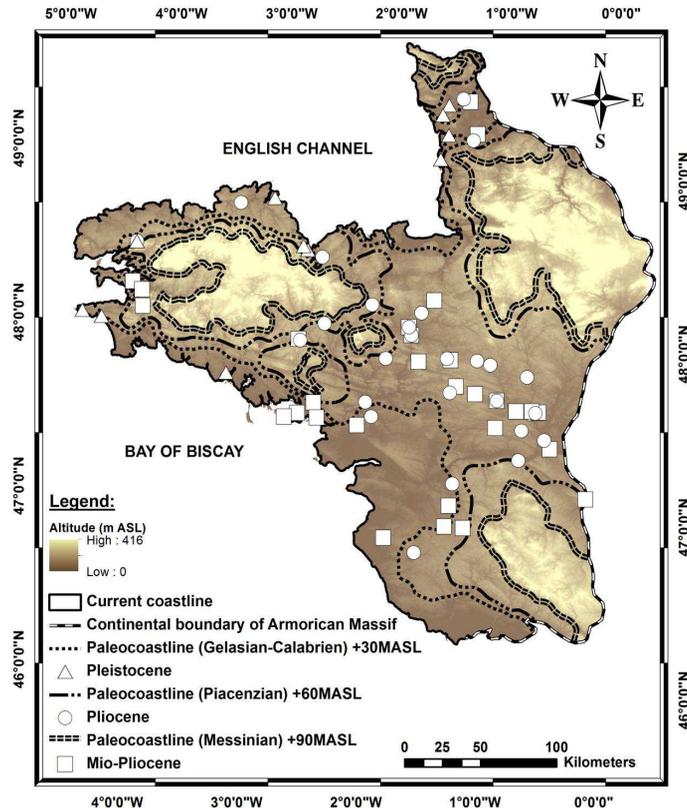


Figure 3. Localization of paleocoastlines on the current topography for past marine transgressions since the Mio-Pliocene time and characteristic sediments associated for each flooding period.

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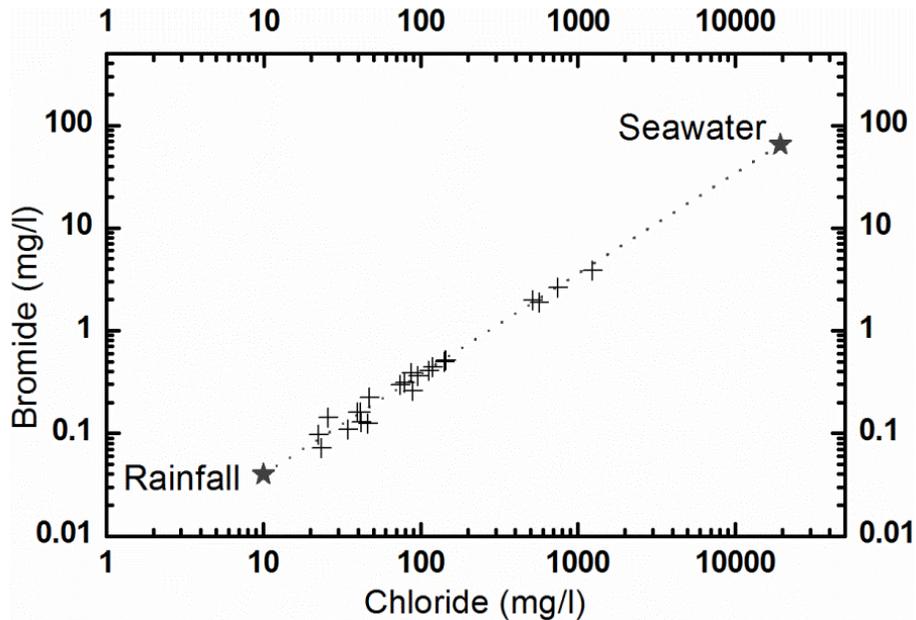


Figure 4. Br vs. Cl concentrations of groundwater in the 12 sites investigated.

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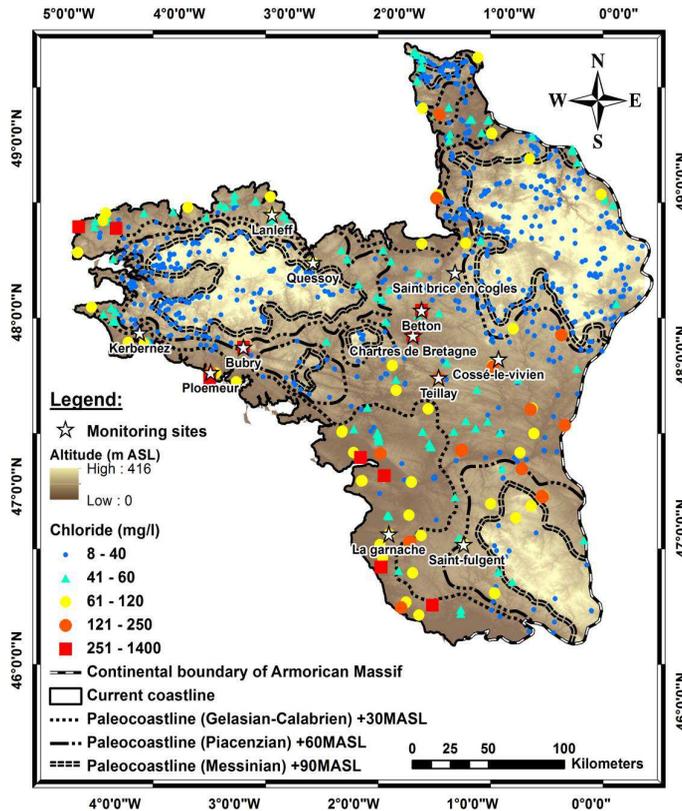


Figure 5. Distribution of paleocoastlines on the current topography for the three transgressions.

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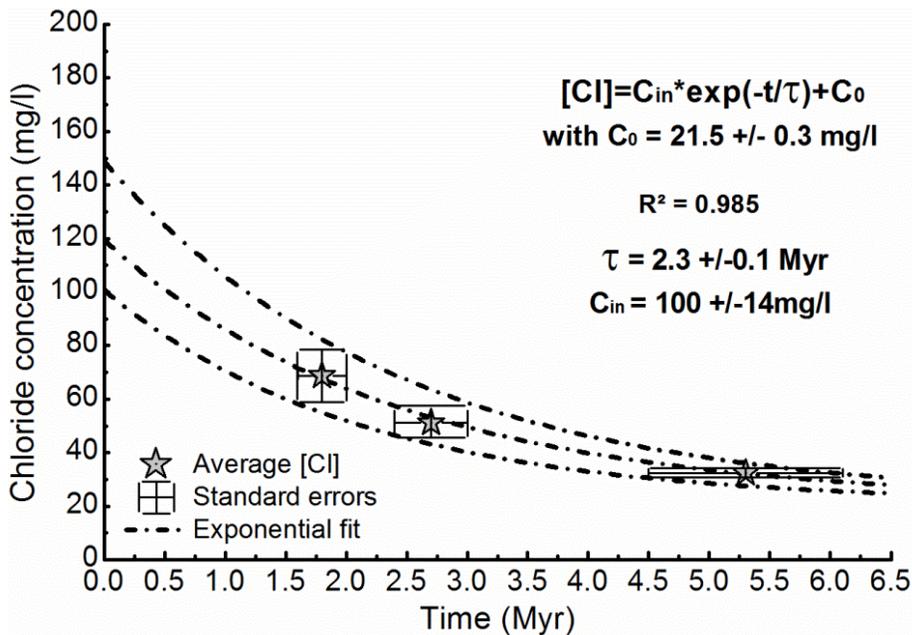


Figure 7. Average chloride concentration for each transgression zone vs. the elapsed time since the transgression.

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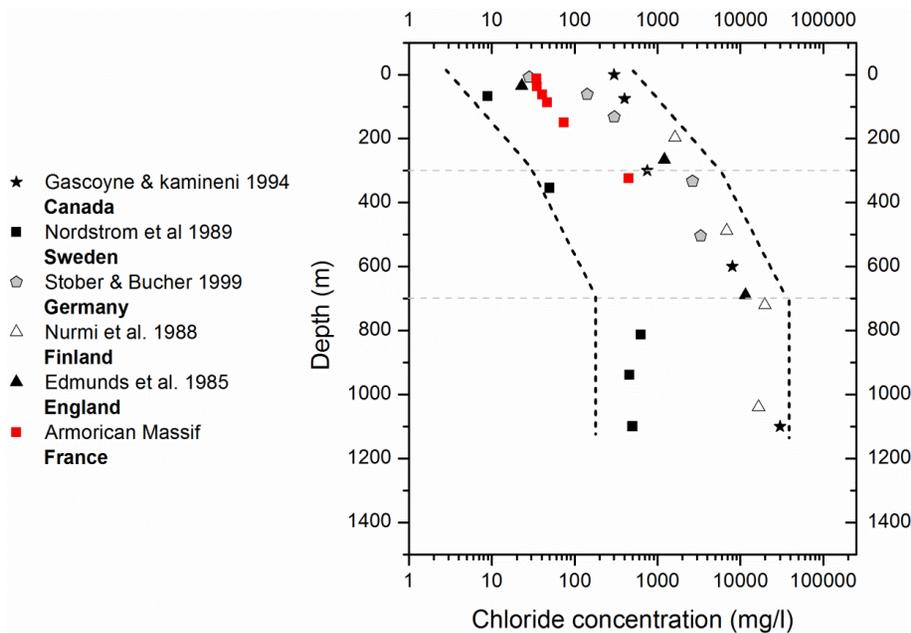


Figure 8. Chloride concentration (mgL^{-1}) vs. depth (m) recorded in continental basement around the world compared with the data of the current study.