



# Quantifying paleorecharge in the Continental Intercalaire (CI) aquifer by a Monte-Carlo inversion approach of $^{36}\text{Cl}/\text{Cl}$ data



J.O. Petersen<sup>a,\*</sup>, P. Deschamps<sup>a,b</sup>, J. Gonçalves<sup>a,c</sup>, B. Hamelin<sup>a,c</sup>, J.L. Michelot<sup>d</sup>, A. Guendouz<sup>e</sup>, K. Zouari<sup>f</sup>

<sup>a</sup> Aix-Marseille Univ., CEREGE, Aix-en-Provence, France

<sup>b</sup> IRD, CEREGE, Aix-en-Provence, France

<sup>c</sup> CNRS, CEREGE, Aix-en-Provence, France

<sup>d</sup> CNRS-UPS, IDES, Orsay, France

<sup>e</sup> University of Blida, Blida, Algeria

<sup>f</sup> ENIS, Sfax, Tunisia

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## ABSTRACT

Past variations of the recharge is estimated in the Atlas Mountains, the main recharge area of the Continental Intercalaire (CI) aquifer, one of the major Saharan aquifers, over the last 775 kyr. In this Mediterranean climatic context, continental archives generally record single humid events, grouped in hypothetical time interval and do not offer continuous chronicles of precipitation. We propose to use spatially distributed  $^{36}\text{Cl}/\text{Cl}$  data as a temporal constraint, to infer the past recharge in the Atlas area. Based on a simplified but robust climatic scenario, assuming a piston model, we apply a Markov Chain Monte Carlo (MCMC) inversion approach to attribute a specific recharge to the last nine interglacial periods and an undifferentiated recharge to the glacial periods. The interglacial recharge values vary from a few  $\text{mm yr}^{-1}$  to more than  $60 \text{ mm yr}^{-1}$ . Glacial recharge is less than  $1 \text{ mm yr}^{-1}$ . These values are then analyzed in terms of intensity and allow questioning some initial hypothesis, especially the generally accepted value of the initial  $^{36}\text{Cl}/\text{Cl}$  value (around  $133 \times 10^{-15}$  at  $\text{at}^{-1}$ ). Our analysis suggests a higher value, around  $175 \times 10^{-15}$  at  $\text{at}^{-1}$ . This approach allows us to bring out paleoclimatic information retained in these continental archives. Especially, computed recharges provide reliable evidence that Marine Isotope Stage (MIS) 5 was more humid than the Holocene period in agreement with marine archives that document wet condition in the North Sahara during that period.

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## 1. Introduction

In a rapidly evolving demographic and climatic environment, groundwater resources management is of strategic importance, especially in arid and semi-arid areas (Taylor et al., 2013). Assessment of the recharge is an essential prerequisite for a sustainable development of the aquifer resource, but remains challenging where the recharge rates are low and variable spatially and temporally. A great deal of effort has been invested to develop different methods for estimating local to regional-scale recharge rates, including physical, chemical, isotopic, and modeling techniques. It has been shown that modern recharge rates are not negligible, albeit limited (see review in Scanlon et al., 2006), contradicting the general viewpoint that much of the groundwater in semi-arid and arid regions is a non-renewable and thus fossil resource. This finding was corroborated by a recent study based on time variation

of total terrestrial water storage inferred from GRACE satellite gravity data (Gonçalves et al., 2013).

However, these approaches are restricted to decadal to century timescales and can only provide a snapshot on the modern history of aquifer recharge. A comprehensive understanding of the general behavior of the large-scale aquifer systems requires extending the investigation of recharge history, and thus to climatic history, to larger timescale (see for example Jost et al., 2007). For such systems, residence time of groundwater in the order of 100,000 years to 1 million years have been revealed by cosmogenic and radio-genic nuclides (e.g.  $^{36}\text{Cl}$ ,  $^4\text{He}$ ,  $^{81}\text{Kr}$ ), in the Great Artesian Basin (GAB) in Australia (Bentley et al., 1986; Phillips et al., 1986; Love et al., 2000; Cartwright et al., 2006; Mahara et al., 2009), the Paris basin in France (Jost et al., 2007; Lavastre et al., 2010; Fourré et al., 2011), the Nubian aquifer in the NE of Sahara (Sturchio et al., 2004; Patterson et al., 2005), or the North Western Saharan Aquifer System, NWSAS (Guendouz and Michelot, 2006). The recharge of these large systems occurred during past, intermittent humid periods within the Quaternary. Provided that they were sufficiently well

\* Corresponding author. Tel.: +33 42971677.

E-mail address: [petersen@cerège.fr](mailto:petersen@cerège.fr) (J.O. Petersen).

dated, these groundwater systems can be considered as first order long-term climatic archives that may have retained evidences for past hydrological and environmental changes (Fontes and Gasse, 1991; Zuppi and Sacchi, 2004; Edmunds et al., 2004). However groundwaters present drawbacks due to dispersion, diffusion and mixing processes that shift and smooth the original geochemical signal (Bethke and Johnson, 2008). Similarly, the numerical groundwater flow modeling, which is now widely used for groundwater resource management, needs to consider the transient nature of the groundwater recharge. Establishment of a reliable time varying recharge scenario is therefore pivotal to successfully simulate “geochemical ages” derived from multiproxy data and fully capture the long-term transient effects on the present-day piezometric trend.

Finally, the interpretation of groundwater dating remains challenging and preferentially requires the use of a multi-proxy.

This study is a first attempt to provide a quantitative reconstruction of the recharge of the Continental Intercalaire (CI), one of the two main aquifers of the NWSAS, over the period including the Holocene to the middle Pleistocene (the last 775 kyr). The computed recharge values are obtained by a coupled approach that combine geochemical tracers, hydrodynamic and transport modeling, as detailed in the following part. Finally, this method allows us to provide quantitative constraints on the intensity of past pluvial periods over the Sahara. The comparison with other paleoclimatic records allows us to discuss the reliability of the overall approach considering the simplifications and assumptions.

## 2. Basic methodology for geochemical data interpretation compared to hydrodynamics

Numerous studies used geochemical tracers including tritium ( $^3\text{H}$ ) and tritium/helium-3 ( $^3\text{H}/^3\text{He}$ ), chlorofluorocarbons (CFCs), carbon-14 ( $^{14}\text{C}$ ), chlorine-36 ( $^{36}\text{Cl}$ ) or chloride mass-balance (CMB) to infer recharge rate of aquifer systems (Cresswell et al., 1999; Scanlon et al., 2002 for review; Alvarado et al., 2005; Cartwright et al., 2006; Wahi et al., 2008; Jiráková et al., 2009; McMahon et al., 2011; Grundl et al., 2013). Some more complex studies combined transport modeling and geochemical tracer distribution either in a crystalline aquifer (Leray et al., 2012) using CFCs or in large-scale sedimentary basins e.g. the Great Artesian Basin, Australia (e.g., Bethke et al., 1999), the Paris basin, France (Castro et al., 1998) using  $^4\text{He}$ , the Gulf coast basin, Texas, USA (Castro and Goblet, 2005), the Black Mesa basin, Arizona, USA (Zhu, 2000) using  $^{14}\text{C}$ , the Nubian Aquifer, Egypt (Patterson et al., 2005) using  $^{36}\text{Cl}$ . These approaches (mainly used for permeability calibration) are generally based on steady-state hydrodynamics and thus involved a constant recharge over time. However, due to large residence times of groundwaters, time-varying recharge conditions are expected. Using a more simple but transient 1D piston-flow model Zhu et al. (2003) estimated the amount of recharge needed to match their chlorine mass balance calculations, supported by  $^{36}\text{Cl}$  data over the last 40 kyr in the Yucca Mountain and Black Mesa aquifer, Arizona, USA. Using MODFLOW simulations, Sanford et al. (2004), after a first set of steady-state hydrodynamic simulations which did not reproduce satisfactorily the  $^{14}\text{C}$  data, made an attempt to simulate the transient-state hydrodynamics by adjusting the recharge over the last 30 kyr in the Rio Grande basin (New Mexico, USA).

In this study, we propose to quantify the time variability of the recharge in a large aquifer using a simplified 1D hydrodynamic and transport model (Piston flow model). Here,  $^{36}\text{Cl}$  data, characterized by a long half-life (301 kyr, against 5.7 kyr for the  $^{14}\text{C}$ ) allows covering a larger period as compared to the previous combined studies outlined above i.e., the last 775 kyr. In addition, a Markov Chain

Monte Carlo (MCMC) inversion approach, not widespread in geochemical studies, was implemented and allows assessing the uncertainty associated with the inversion process of the recharge history. To summarize, this work is based on a set of four approaches presented in Fig. 1 (see also Section 5). A simple piston flow model simulates the  $^{36}\text{Cl}$  data, in accordance with two climatic scenarios involving two kinds of recharge, one corresponding to humid episodes (in blue in Fig. 1) and one corresponding to dry episodes (in green in Fig. 1). Therefore, the amount of water recharging the CI and the distance travelled by the groundwater is time-dependent. Assuming piston flow, Fig. 1 illustrates that large distances are covered by groundwaters during humid periods and conversely weak distances are travelled during dry periods. Consequently, this alternation is translated into the  $^{36}\text{Cl}$  signal with radioactive decay occurring over large and weak distances during humid and dry periods, respectively (Fig. 1C).

After a brief presentation of the hydrogeological context (see Section 3), geochemical and isotopic ( $^{36}\text{Cl}$ ) data that document streamlines in the North of the CI are presented in Section 4. Then, the mathematical model i.e., the piston flow model based on suitable climatic scenarios and the MCMC approach are presented (see Section 5). Finally we present the results and discuss them in the light of the paleoclimatic literature.

## 3. Background

### 3.1. Geological and hydrogeological settings

The NWSAS is one of the major transboundary aquifer system in the world, with a surface extent of more than  $10^6 \text{ km}^2$ , and a depth range of up to 3 km. It is constituted predominantly by Triassic to Quaternary continental formations, that include two main aquifer layers: the “Complex Terminal” (CT) and “Continental Intercalaire”

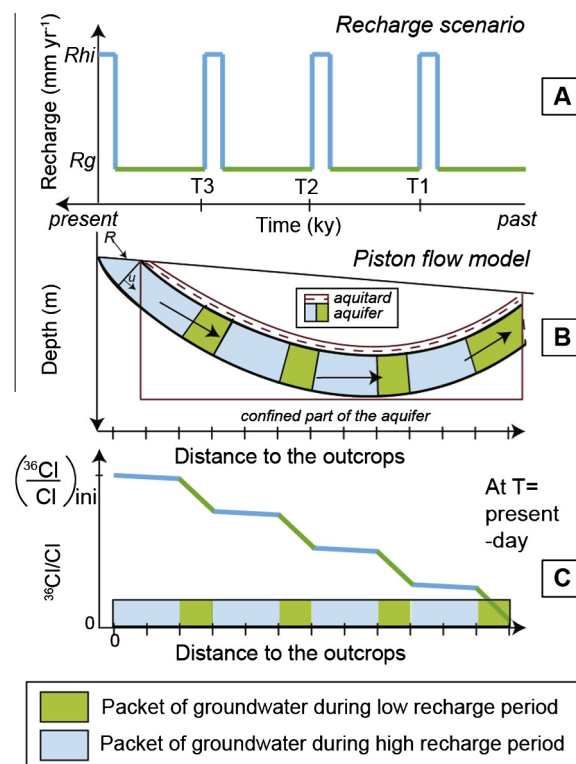


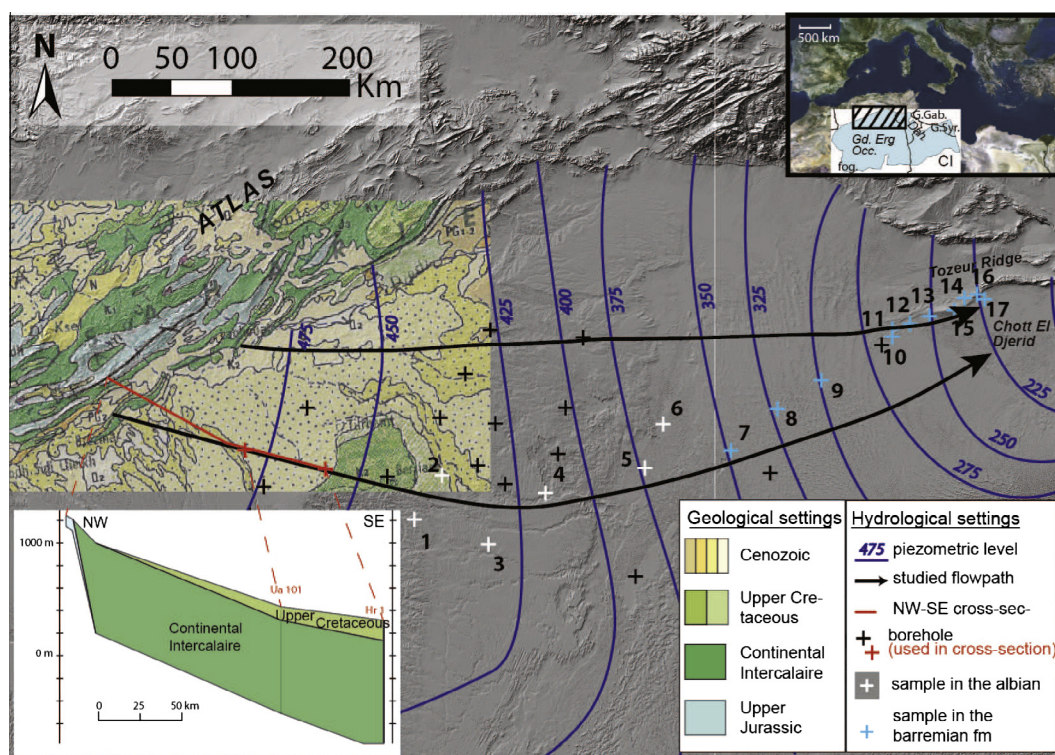
Fig. 1. Piston flow model (B) and resulting  $^{36}\text{Cl}/\text{Cl}$  distribution (C) in a confined aquifer as a function of distance to the outcrops and considering an alternation of arid and humid recharge period (A).

(CI). The CT, shallow and mostly unconfined, integrates upper Cretaceous to Miocene layers. In this study, we focus on the deeper and mostly confined CI aquifer, constituted by middle Jurassic to lower Cretaceous formations described as heterogeneous sandy units with variable clay content, 350 m thick in average (Baba Sy, 2005 and the references therein). Hydraulic and geochemical data reveal the continuity of the main flowlines (Edmunds et al., 2003). Outcrops, in the Atlas Mountains in Algeria and Dahar Mountains in Tunisia, and unconfined areas in a major part of the Grand Erg Occidental, represent about 20% of the total surface extent of the CI (Fig. 2). The main discharge areas are the Tunisian Gulf of Gabes, the Libyan Gulf of Syrte and the Algerian Foggaras, in the SW of the Grand Erg Occidental. Estimates of the present-day recharge rate based on hydrogeological models range between 5.6 and 11.9 m<sup>3</sup> s<sup>-1</sup> (Baba Sy, 2005 and references therein) corresponding to an annual recharge between 6 and 12 mm. Baba Sy (2005) also estimated a paleo-recharge around 29 m<sup>3</sup> s<sup>-1</sup> at the beginning of the Holocene. In a recent study based on the GRACE satellite record of terrestrial water storage for the period 2003–2010, Gonçalves et al. (2013) estimated a cumulative recharge rate of 44 ± 28 m<sup>3</sup> s<sup>-1</sup> in the multilayered NWSAS, corresponding to 2.1 ± 1.3 mm yr<sup>-1</sup> for the CI itself. In the present study, we investigate the hydrological and geochemical variations along a longitudinal transect in the northern part of the CI, from the east of the Atlas Mountains in Algeria, to the Tozeur Ridge bordering the north of Chott El Djerid in Tunisia. The general eastward circulation of the CI groundwater along this transect is illustrated in Fig. 2 by two streamlines inferred from the modern piezometric map (OSS, 2003), bracketing the position of our samples. Information about the lithology, depth, and thickness of the layers bearing the CI is available from more than twenty borehole logs in the region (ERESS, 1972; Fig. 2). The most hydraulically efficient levels are the Barremian, Albian and Neocomian (Hauterivian and

Valanginian stages) sandstone formations. The average total thickness is 800 m. The Barremian and Albian formations are separated by 7 to 30 m of Aptian dolomite. Porosity measurements are scarce: values between 22% and 26% have been reported in the Algerian part of the CI, mainly in the Albian formation (ERESS, 1972), and between 18% and 21% in the Barremian formation in the Chott El Djerid region (Guellala et al., 2011). A general trend of increasing clay content is observed in the sandstone along the transect, with clay proportion ranging from 20% to 70%. Interpretation of the stratigraphic logs indicates that the CI is confined by 300 to 1500 m of overlying formations over most of its surface. Close to the outcrop, Upper Jurassic and Lower Cretaceous outcrops appear as a SW–NE folded and faulted belt. Seismic data (Bracene and de Lamotte, 2002), as well as examination of the geological map (Fig. 2) suggest that the width of the unconfined area is comprised between 10 and 20 km, in good agreement with the estimate of Baba Sy (2005).

### 3.2. Climatic context

Today northwestern Africa is dominated by two climate systems, the Mediterranean and the northwest African monsoonal climate systems, separated by the Saharan desert belt, the modern location of which extends between ~20–24°N (Nicholson, 2000). Extending from the northern edge of the Saharan belt to the Atlas Mountains, NWSAS is located in the strongly seasonal Mediterranean climate with winter rainfalls and dry summer (Nicholson, 2000). Quantitative estimations of the precipitation are spatially continuous and modeled from satellite observations of clouds or scarce and directly measured using pluviometers. Satellite-derived rainfall rates are available at a resolution of 1° × 1° on most of the globe since 1979 ([http://gdml1.sci.gsfc.nasa.gov/daac-bin/G3/gui.cgi?instance\\_id=GLDAS10\\_M](http://gdml1.sci.gsfc.nasa.gov/daac-bin/G3/gui.cgi?instance_id=GLDAS10_M)). They indicate annual rainfall



**Fig. 2.** Studied area, between Algeria and Tunisia. This figure shows the hydrological settings, the piezometric level (m), a cross-section modified from Baba Sy (2005), the location of boreholes used to described the lithology and the wells of the studied samples. Geological details (from the CCGM geological map of Africa, 1:1,000,000 scale) indicate the CI outcrops. The background is a SRTM provided by ESRI®. The inset shows the CI in the mediterranean context and key places for the recharge: Dahar Mountains (Dah.) and the discharge: Gulf of Syrte (G. Syr.), Gulf of Gabes (G. Gab) and the foggaras (fog.) in the Grand Erg Occidental (Gd. Erg Occ.).

rate around 200 mm yr<sup>-1</sup>. The precipitations measured since the 1970s and reported in OSS (2003) and Baba Sy (2005) correspond to an annual mean between 80 and 350 mm on the Atlas Mountains area. In both cases, the maximum rates are observed between October and May. This large range emphasizes the orographic gradient according to the SW–NE orientation of the Atlas Mountain. Rainfall rate are maximum in the northwestern flank of the Atlas Mountains and decrease southward this topographic barrier, while the southern part of NWSAS experiences episodic rainfall (annual mean <10 mm). Precipitation over the NWSAS area results from the combined influences of the Atlantic Ocean, the Mediterranean Sea and the Sahara belt, together with very steep orography of the Atlas region (Knippertz et al., 2003; Trambly et al., 2012). Isotopic composition of modern precipitation highlights Atlantic source of humidity over western Mediterranean and its African fringe (Sonntag et al., 1979; Sultan et al., 1997; Abouelmagd et al., 2012). During the Quaternary, marine and terrestrial records testify of important climatic and hydrological changes in North Africa and in the Mediterranean realm. It is not the aim of this article to exhaustively review the numerous studies that have documented these fluctuations. Comprehensive reviews of climatic and environmental changes over these zones and that encompass the NWSAS region can be found in Gasse (2000), Hoelzmann et al. (2004) and Abrantes et al. (2012). The general picture emerging from these syntheses of paleoenvironmental records from Northern Africa and the surrounding seas (north of 10°N) is that long-term changes in the magnitude of pluvial periods have been mainly paced by Milankovitch cycles. Although secondary modulations in the hydrological cycle can be superimposed to these orbitally-forced climate variations, interglacial periods, including the Holocene, were characterized by wet conditions over the recharge area of the NWSAS and were responsible for active recharge of this aquifer system. Conversely, glacial periods, especially glacial maxima were much drier. This general pattern, well-documented for the last glacial–interglacial alternation can be reasonably extended to the 100,000 year cycles that characterized earth climate over the late Quaternary. This is in general agreement with U–Th dating of authigenic carbonate deposits that has allow to correlate Saharan paleolake occurrences with major interglacial stages MIS 9, MIS 7, MIS 5e, and MIS 1 (MIS: Marine Isotope Stage) at least the last 400 kyr, with pluvial periods centered to interglacials (Causse et al., 2003; Geyh and Thiedig, 2008; Sultan et al., 2007; Szabo et al., 1995; Szabo et al., 1989).

#### 4. Geochemical data and groundwater <sup>36</sup>Cl ages

Chronological constraints along the investigated area in this study rely on seventeen <sup>36</sup>Cl data. Nine of them, located in Algeria were initially published in Guendouz and Michelot (2006). Eight others (see Table 1) are located along the Tozeur Ridge in Tunisia, on the easternmost part of the studied area. They are part of a more global study of the Tunisian part of the CI aquifer and will be presented in more details elsewhere. The global geochemical context of the northern part of the CI is detailed in Edmunds et al. (2003) and Guendouz and Michelot (2006).

##### 4.1. Geochemical data

The chloride content and <sup>36</sup>Cl/Cl ratios discussed in this study are illustrated in the longitudinal profile shown in Fig. 3, and our new data are listed in Table 1. Chloride content varies from 188 mg l<sup>-1</sup> close to the CI outcrops in the Atlas Mountains, up to 560 mg l<sup>-1</sup> in sample #7, #8 and #9 close to the Algerian border in the Barremian layer. The general increase eastward along the transect, as well as the Br/Cl and I/Cl ratios were interpreted by

**Table 1**

Chloride content (mg l<sup>-1</sup>) content, <sup>36</sup>Cl/Cl ratio (<sup>36</sup>R × 10<sup>-15</sup> at at<sup>-1</sup>) and associated <sup>36</sup>R<sub>cor</sub> for the 17 samples considered in this study. Data for samples (#1 to #9) are from Guendouz and Michelot (2006). The size of the correction of evaporite dissolution (Cor%) is expressed in %. The distance from the outcrop (D) is given in km.

| Samp. | Long.<br>(dd) | Lat.<br>(dd) | D<br>(km) | Cl<br>(mg l <sup>-1</sup> ) | <sup>36</sup> R ± (× 10 <sup>-15</sup> at at <sup>-1</sup> ) | <sup>36</sup> R <sub>cor</sub> | Cor%<br>(%) |
|-------|---------------|--------------|-----------|-----------------------------|--------------------------------------------------------------|--------------------------------|-------------|
| 1     | 3.9968        | 32.5845      | 225       | 210                         | 99 34                                                        | 104                            | 5           |
| 2     | 4.2059        | 32.8745      | 242       | 188                         | 95 8                                                         | 95                             | 0           |
| 3     | 4.5506        | 32.4134      | 276       | 511                         | 29 8                                                         | 74                             | 61          |
| 4     | 5.0189        | 32.7081      | 321       | 550                         | 18 8                                                         | 50                             | 64          |
| 5     | 5.7177        | 32.8912      | 390       | 533                         | 10 5                                                         | 27                             | 62          |
| 6     | 5.9008        | 33.1717      | 417       | 426                         | 15 5                                                         | 32                             | 53          |
| 7     | 6.3667        | 32.9934      | 459       | 560                         | 23 3                                                         | 64                             | 64          |
| 8     | 6.7470        | 33.3048      | 500       | 480                         | 25 3                                                         | 60                             | 58          |
| 9     | 7.0608        | 33.4807      | 544       | 426                         | 14 3                                                         | 29                             | 53          |
| 10    | 7.5811        | 33.8192      | 472       | 270                         | 33 2                                                         | 44                             | 26          |
| 11    | 7.5939        | 33.7514      | 474       | 359                         | 29 2                                                         | 53                             | 44          |
| 12    | 7.7225        | 33.8358      | 487       | 290                         | 29 2                                                         | 42                             | 31          |
| 13    | 7.8719        | 33.8875      | 501       | 296                         | 25 2                                                         | 38                             | 32          |
| 14    | 8.1925        | 34.0017      | 527       | 293                         | 9 1                                                          | 13                             | 32          |
| 15    | 8.2453        | 34.0194      | 537       | 416                         | 31 2                                                         | 65                             | 52          |
| 16    | 8.2619        | 34.0150      | 539       | 414                         | 34 2                                                         | 70                             | 52          |
| 17    | 8.1139        | 33.9119      | 523       | 536                         | 15 1                                                         | 40                             | 63          |

Edmunds et al. (2003) as evidence of progressive dissolution of non-marine evaporites along the flow lines. Samples from the Barremian level located in the northeastern part of the study area are in the same range of concentration (270–536 mg l<sup>-1</sup>). Together, these samples are thus in a diluted range of concentration, compared to other regions of the CI aquifer where concentrations up to several g l<sup>-1</sup> are quite common.

<sup>36</sup>Cl/Cl ratios strongly decrease eastward from 99 ± 34 to 10 ± 5 × 10<sup>-15</sup> at at<sup>-1</sup> along the transect in the Albian level in Algeria. There is a sharp fall between sample #2 and #3, corresponding to a parallel increase in [Cl<sup>-</sup>] concentration. All the samples to the east, in the Barremian samples both in Algeria and Tunisia, are confined within a range of low ratios between 33.8 ± 2.1 × 10<sup>-15</sup> and 8.8 ± 1.03 × 10<sup>-15</sup> at at<sup>-1</sup>, with no clear spatial repartition.

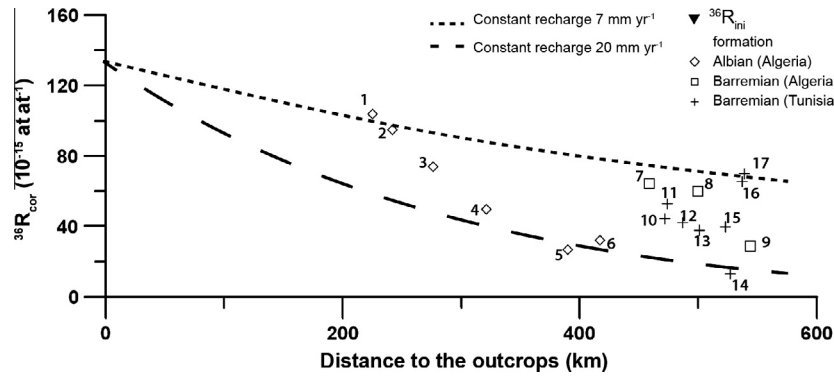
##### 4.2. Chronological constraints derived from <sup>36</sup>Cl data

Reliable groundwater residence time values can be derived from <sup>36</sup>Cl data only once several intrinsic limitations have been considered (Phillips et al., 1986; Davis et al., 1998; Love et al., 2000). First, the input values of <sup>36</sup>Cl content and <sup>36</sup>Cl/Cl ratio in recharge water must be properly assessed. While the isotopic ratio is insensitive to the huge concentration effect due to evaporation under arid climate, it may be heavily impacted by either <sup>36</sup>Cl or Cl addition along the groundwater flowpath. The most significant interfering processes are (1) chloride supply from evaporite dissolution, (2) subsurface (*in situ*) production of <sup>36</sup>Cl, (3) diffusion from neighbor aquitards, or mixing with adjacent aquifers.

The budget equation of <sup>36</sup>Cl is written classically as:

$$^{36}\text{Cl} = ^{36}\text{R} \times \text{Cl} \\ = ^{36}\text{R}_i \times \text{Cl}_i \times e^{-\lambda t} + ^{36}\text{R}_e \times \text{Cl}_i \times (1 - e^{-\lambda t}) + ^{36}\text{R}_s(\text{Cl} - \text{Cl}_i) \quad (1)$$

where  $\lambda$  is the decay constant of <sup>36</sup>Cl (2.303 × 10<sup>-6</sup> yr<sup>-1</sup>),  $t$  is the time elapsed since the recharge (s), <sup>36</sup>Cl the concentration (atoms l<sup>-1</sup>), <sup>36</sup>R the measured <sup>36</sup>Cl/Cl ratio, Cl the measured chloride concentration of the sample (atoms l<sup>-1</sup>), at time  $t$ , Cl<sub>i</sub> the initial chloride concentration (atoms l<sup>-1</sup>), <sup>36</sup>R<sub>i</sub> the initial <sup>36</sup>Cl/Cl ratio, <sup>36</sup>R<sub>e</sub> the secular equilibrium <sup>36</sup>Cl/Cl ratio in the aquifer and <sup>36</sup>R<sub>s</sub> the secular equilibrium <sup>36</sup>Cl/Cl ratio of non-meteoritic chloride added to the system by diffusion or mixing. Secular equilibrium is reached when radioactive decay of meteoric <sup>36</sup>Cl and *in situ* production of <sup>36</sup>Cl are balanced.



**Fig. 3.**  $^{36}\text{R}_{\text{cor}}$  ratio (corrected for evaporite dissolution) of the studied samples as a function of the distance to the outcrops, compared with the theoretical evolution of the  $^{36}\text{R}_{\text{cor}}$  ratio assuming steady-state conditions. Are presented two scenarios with constant recharge rate of  $7 \text{ mm a}^{-1}$  and  $20 \text{ mm a}^{-1}$ .

The initial  $^{36}\text{R}_i$  ratio can be constrained in different ways (Davis et al., 1998). Based on a theoretical value of the natural  $^{36}\text{Cl}$  fallout rate at the Saharan Atlas latitude ( $\sim 34^\circ\text{N}$ ) (Lal and Peters, 1967; Parrat et al., 1996), combined with the average chloride content in precipitation, Guendouz and Michelot (2006) calculated a first estimate of  $116 \times 10^{-15} \text{ at at}^{-1}$ . These authors also performed direct measurements in leached soil samples collected in the CI recharge area, that gave an average  $^{36}\text{R}_i$  of  $133 \times 10^{-15} \text{ at at}^{-1}$ , consistent with the value of  $131 \times 10^{-15} \text{ at at}^{-1}$  estimated by Patterson et al. (2005) for the Nubian Aquifer. Recent data obtained in the Tunisian CT range up to  $118 \times 10^{-15} \text{ at at}^{-1}$  (associated to chloride concentration around  $400 \text{ mg l}^{-1}$ , Hadj Ammar, pers. com.). We first use the upper bound value ( $133 \times 10^{-15} \text{ at at}^{-1}$ ) as an input in the following calculations. Secular variations of the  $^{36}\text{Cl}$  production rate related to fluctuations of the geomagnetic field strength over long timescales (e.g. Baumgartner et al., 1998; Muscheler et al., 2005) as well as changes in precipitation sources and regime may have affected differently  $^{36}\text{Cl}$  and  $\text{Cl}$  and may likely result in variation over time of this key parameter. Although it is difficult to assess *a priori* this fluctuation over the considered timeframe, the sensitivity of our model results to such fluctuations will be discussed further below. Other different processes listed above, that affect  $^{36}\text{Cl}/\text{Cl}$  along the underground transit in the aquifer were already discussed in detail in previous studies. We assume that diffusion from adjacent aquitards is likely negligible, due to the large thickness of the CI aquifer (Bethke and Johnson, 2008).

Available major and trace element data, seem to indicate that no mixing process with other aquifers occurs along the flowpaths investigated in this study (Edmunds et al., 2003; Guendouz and Michelot, 2006). The last term in Eq. (1) is thus neglected. Following Patterson et al. (2005) we adopt a secular equilibrium value  $^{36}\text{R}_e$  of  $5 \times 10^{-15} \text{ at at}^{-1}$  for the *in situ* production of  $^{36}\text{Cl}$  by thermal neutron capture ( $^{35}\text{Cl}(n, \gamma)^{36}\text{Cl}$ ) in the aquifer (Fabryka-Martin et al., 1987; Lehmann et al., 2003). As mentioned above, there is strong evidence that the main perturbation results from chloride addition along the flowpath, due to evaporites dissolution. We can thus correct the measured isotopic ratio for this contamination, assuming again that evaporites release dead chlorine devoid of  $^{36}\text{Cl}$ .

Following this assumption, Eq. (1) then becomes:

$$^{36}\text{R}_{\text{cor}} = ^{36}\text{R} \times \frac{\text{Cl}}{\text{Cl}_i} = ^{36}\text{R}_i \times e^{-\lambda t} + ^{36}\text{R}_e \times (1 - e^{-\lambda t}) \quad (2)$$

where  $^{36}\text{R}_{\text{cor}}$  is the “corrected” ratio only affected by radioactive decay and *in situ* production. The initial chlorine concentration ( $\text{Cl}_i$ ) is taken as  $200 \text{ mg l}^{-1}$  as a mean of chlorine concentration observed in the recharge area (Guendouz and Michelot, 2006). The size of the correction is listed in Table 1, and ranges from 4%

to 70% of the measured ratio.  $^{36}\text{R}_{\text{cor}}$  now varies from 16 to  $130 \times 10^{-15}$ .

## 5. Modeling approaches

### 5.1. Simulating $^{36}\text{Cl}$ data spatial distribution using a piston flow model

Piston flow is based on the simple assumption that groundwater moves along a flowpath from the recharge area towards the discharge area as a isolated packet of water without exchanging water molecules with adjacent packets or neighboring aquitards (Bethke and Johnson, 2008). This simple but robust flow model may be suitable to describe parts of large and homogeneous confined aquifers and have been successfully applied to the Great Artesian Basin or the Nubian aquifer (Dincer et al., 1974; Cook et al., 1992; Cook and Böhlke, 2000; Scanlon et al., 2002; Herczeg and Leaney, 2011). Following the reasoning of piston flow model, the pore fluid velocity  $u$  resulting from a mass balance equation writes:

$$u = \frac{R(t)}{\omega} \times \frac{L}{l} \quad (3)$$

where  $R$  ( $\text{mm yr}^{-1}$ ) is the annual recharge,  $\omega$  the porosity,  $L$  (m) the length of the outcrops, and  $l$  (m) the aquifer thickness (Cook and Böhlke, 2000; Scanlon et al., 2002). Then, the distance  $\mathbf{X}(\tau)$  travelled by a packet of groundwater from a recharge point at  $t = -\tau$  to its present position ( $t = 0$ ), is:

$$\mathbf{X}(\tau) = \int_{-\tau}^0 \mathbf{u}(\mathbf{t}) \times d\mathbf{t} \quad (4)$$

By definition, a piston flow model does not reproduce the transient state occurring at the transition between low and high recharge periods. Therefore an alternation of over- and underestimations can be expected due to the lag time between the recharge variation occurrence and the effective new steady state. Rousseau-Gueutin et al. (2013) calculated time necessary to reach a near-steady state after a variation of the recharge for several large aquifers in the world. This time ranges between 0.7 and  $2.4 \times 10^7 \text{ kyr}$  depending on the physical properties of the system (storativity, transmissivity, and length of the aquifer). Assuming that the CI is mostly confined, using the average value of the transmissivity ( $T \approx 10^{-2} \text{ m}^2 \text{ s}$ ) and the specific storage ( $S \approx 10^{-3}$ ) given in OSS (2003), yields a hydraulic diffusivity  $\text{Dh} = \frac{T}{S} \approx 10$  and a characteristic time  $\tau = L^2/\text{Dh} \approx 500,000^2/10 = 2.5 \times 10^{10} \text{ s} \approx 700 \text{ yr}$ . Consequently, if  $R$  becomes zero, in the CI, the associated steady state ( $u = 0$ ) is reached after about  $3\tau \approx 2000 \text{ a}$ , which is very low compared to the 775 kyr simulation time (or more rigorously to the 323 kyr of the mainly recharging interglacial periods) and thus

almost instantaneous. Besides its simplicity, the major interest of such a model is that the knowledge of the hydraulic conductivity is not needed.

Piezometric level showed in Fig. 2 indicates divergent then parallel flowpaths. In the easternmost section, they become slightly convergent but the mixing zone of the discharge area of the Gulf of Gabes is not reached. Consequently, the piston flow model can be considered suitable to model the flow regime in the northwest of the CI. As described in Section 3.1, we retain a thickness  $l$  of 800 m and an outcrop length  $L$  of about 15 km. A simple geometric calculation shows that the ratio  $L/l$  that appears in Eq. (3) is independent from  $l$  and depends only on the mean geological dip of the geological formation. In addition, for monoclinic formations, the same ratio is valid for the sub-units within a geological layer. Therefore the available ratio for the CI can be used for both the Albian and the Barremian formations. The  $^{36}\text{Cl}$  distribution within the CI aquifer can then be simulated in a simple form. A theoretical  $^{36}\text{Cl}/\text{Cl}$  ratio, noted  $^{36}\text{R}_{\text{Th}}$ , can be calculated for any time  $t = \tau$  from Eq. (2):

$$^{36}\text{R}_{\text{Th}} = ^{36}\text{R}_i \times e^{-\lambda\tau} + ^{36}\text{R}_{\text{eq}} \times (1 - e^{-\lambda\tau}) \quad (5)$$

Combination of Eqs. (3)–(5) fully describes the distribution of  $^{36}\text{R}$  along a flow line, for a system without dissolution or diffusion. Indeed, for a given time  $t = -\tau$ , the migration distance  $X$  for a volume of groundwater that recharged the confined aquifer at that time can be computed. Therefore, for any recharge scenario  $R(t)$ , it is possible to simulate  $^{36}\text{R}_{\text{Th}}$  distribution along a flowpath and directly compare these simulated values to  $^{36}\text{R}_{\text{cor}}$  distribution. Using the geometrical characteristics indicated above for the CI outcrop ( $l = 800$  m and  $L = 15$  km) and assuming a mean porosity of 20%, a first set of  $^{36}\text{R}_{\text{Th}}$  simulations can be performed under the simplifying assumption of a time constant recharge. The  $^{36}\text{R}_{\text{cor}}$  data considered in this study and the simulated  $^{36}\text{R}_{\text{Th}}$  values are plotted in Fig. 3 against the distance from the outcrop.

Two sets of  $^{36}\text{R}_{\text{Th}}$  values are presented in Fig. 3, considering two alternative values for the recharge, one representative of present-day condition ( $7 \text{ mm yr}^{-1}$ ), and the other of the Holocene humid period ( $20 \text{ mm yr}^{-1}$ ).

Two important insights can be drawn from the poor agreement between measured and simulated data illustrated in Fig. 3. First, the different trends observed by the Barremian and the Albian data suggest a higher pore fluid velocity in the Barremian formation, i.e. similar  $^{36}\text{R}_{\text{cor}}$  values at higher distance from the recharge area. A difference in the ratio  $L/l$  (see Eq. (4)) is unlikely to explain these contrasted velocities since some of the Barremian samples belong to the same streamline than the Albian samples in a monoclinic configuration. A more plausible explanation for this difference is the confirmation of the higher porosity reported in the Albian formation (ERESS, 1972; Guellala et al., 2011, Section 3.1). Therefore, in the following simulations, we will use porosity values of 26% and 19% for the Albian and Barremian formations, respectively, (i.e. the highest and lowest bound of the measured range).

More importantly, this first simulation clearly highlights that the assumption of constant recharge is unrealistic and that usual steady-state models cannot fit the distribution of geochemical tracers. Recharge scenarios similar to those schematically depicted in Fig. 1 are tested in the next sections in order to improve the simulations.

## 5.2. Global climatic scenario and recharge constraints

The general pattern of dry-glacial and wet-interglacial periods has been well documented for the last glacial–interglacial alternation in Northern Africa (see Section 3.2). Then we define two general scenarios of recharge variations over that period as defined in Fig. 4. The first scenario is based on square-wave alternation between a

maximum recharge rate ( $\text{Rh}_i$ ) during interglacial and the low recharge rate ( $\text{Rg}$ ) during glacial periods. The second scenario is a saw-toothed waveform characterized by abrupt deglacial transitions from full-glacial recharge ( $\text{Rg}$ ) to wet interglacial conditions ( $\text{Rh}_i$ ), followed by a much more gradual return into the dry glacial conditions. Similar simplified schemes have often been used to picture climatic proxies such as benthic forams  $\delta^{18}\text{O}$  or atmospheric  $\text{CO}_2$  over the last ice-age cycles (Ruddiman, 2003; Ruddiman, 2006).

According to the first  $^{36}\text{Cl}$ -ages estimated by Guendouz and Michelot (2006), ages of more than 700 kyr can be expected for the CI groundwater along the investigated transect, implying that nine glacial cycles must be considered in our scenarios. Timing and duration of glacial and interglacial periods are given by the age model of the  $\delta^{18}\text{O}$  benthic stack (“LR04”) of Lisiecki et al. (2005), established by tuning the marine  $\delta^{18}\text{O}$  record to insolation. Transitions between glacial and interglacial periods are abrupt, making the timing of their inception easy to determine (Fig. 4). For the four latter terminations, absolute chronological constraints provided by Cheng et al. (2009) are used. By contrast, defining the end of the full-interglacial period is more difficult. We arbitrarily fix the date of the terminations by setting the limit of full-interglacial condition at around 4‰ on the  $\delta^{18}\text{O}$  marine record (Fig. 4).

Past recharge *a priori* values ( $\text{Rh}_i$ ) are needed as inputs to the inversion process and Bayesian approach exposed below. Considering a present-day precipitation of about  $250 \text{ mm yr}^{-1}$  in the Atlas Mountains, and a recharge rate of between 6 and  $12 \text{ mm yr}^{-1}$  in the CI (Baba Sy, 2005), yields a deep recharge/rainfall ratio of 3–5%. Similar values were found for large confined aquifers in different climatic contexts: 4% was reported for the Albian aquifer in the temperate Paris basin (Raoult, 1999), and a ratio of 1–3% for the semi-arid Great Artesian basin. Using a realistic range for deep recharge to rainfall ratio of 1–5% and the estimation of precipitation of around  $600 \text{ mm yr}^{-1}$  during the Holocene period (Desprat et al., 2013), we obtain a recharge rate within  $6\text{--}30 \text{ mm yr}^{-1}$  during the Holocene. In addition, we assume a maximum tenable rainfall of  $2000 \text{ mm yr}^{-1}$  in the Atlas Mountains, corresponding to a maximum recharge rate of about  $100 \text{ mm yr}^{-1}$  for the other humid periods.

## 5.3. Markov Chain Monte Carlo inversion approach

Once the recharge scenario is defined, the recharge values, considered as parameters of the system, can be constrained using an

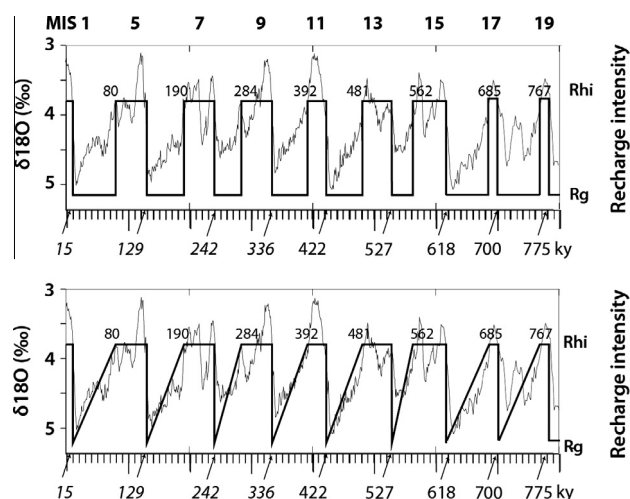


Fig. 4. Square wave (top panel) and saw-tooth (bottom panel) recharge scenarios considered in this study. Pluvial periods were established according to the  $\delta^{18}\text{O}$  benthic stack provided by Lisiecki et al. (2005), with a lower limit around 4‰. The recharge intensity scale is arbitrary.

inverse approach based on  $^{36}\text{Cl}$  data ( $^{36}\text{R}_{\text{cor}}$ ) and modeled  $^{36}\text{Cl}$  ( $^{36}\text{R}_{\text{Th}}$ ) distribution (see Fig. 2). Here, the data set comprises 6 and 11 data for the Albian and the Barremian formation, respectively. For parsimony reasons but also to overcome under-determination problems in the inverse process, 9 interglacial period recharge values ( $\text{Rh}_i = 1, \dots, 9$ ), and a unique glacial period recharge  $\text{Rg}$  are considered in the piston-flow model. Such specific focus on the interglacial periods is based on the fact that the CI aquifer was mostly recharged during the humid periods while glacial periods contribution is limited to a few  $\text{mm yr}^{-1}$ . So defined, our inverse problem thus involves 10 parameters and 17  $^{36}\text{Cl}$  data. Inverse methods imply an automatic procedure to constraint model parameters in order to reduce the misfit between calculated and measured values (see e.g. Tarantola, 2005, chap. 2 for methods). From a general standpoint, the misfit function between simulated ( $\text{sim}_i$ ) and measured ( $\text{obs}_i$ ) variables can be written as:

$$S(\mathbf{m}) = \sum_i \frac{|\text{obs}_i - \text{sim}_i|}{\sigma_i} \quad (6)$$

where  $\sigma_i$  is the uncertainty on each measurement, and  $\mathbf{m}(m_1, \dots, m_n)$  the vector of model parameters. Since the piston-flow model computes both distance ( $X_i$ ) and  $^{36}\text{R}_{\text{Th}}$ , the misfit function should involve the two variables. In addition, in order to give a similar weight to the Albian and the Barremian samples, one term for each formation is introduced. This more exhaustive misfit function used here in the MCMC method thus writes:

$$S(\mathbf{m}) = \sum_{i=1}^6 \frac{|^{36}\text{R}_{\text{cor}_i} - ^{36}\text{R}_{\text{Th}_i}|}{\sigma_i} \bigg/ \sum_{i=1}^6 \frac{1}{\sigma_i} \bigg|_{\text{Alb}} + \sum_{i=1}^{11} \frac{|^{36}\text{R}_{\text{cor}_i} - ^{36}\text{R}_{\text{Th}_i}|}{\sigma_i} \bigg/ \sum_{i=1}^{11} \frac{1}{\sigma_i} \bigg|_{\text{Bar}} + \frac{1}{6} \sum_{i=1}^6 |X_i^{\text{sim}} - X_i^{\text{obs}}|_{\text{Alb}} + \frac{1}{11} \sum_{i=1}^{11} |X_i^{\text{sim}} - X_i^{\text{obs}}|_{\text{Bar}} \quad (7)$$

From a theoretical standpoint, inverse methods explore how observations can bring valuable information on model parameterization. Among the available methods, one can distinguish deterministic and probabilistic (or “Bayesian”) inverse methods. Deterministic approaches such as the gradient methods look for a unique parameter set that allows the best fit between the model and the data. Although attractive, the existence of a unique solution underlain by deterministic inverse methods disregards a major feature in inverse modeling i.e., a possible large number of parameter sets which can lead to almost equally satisfying simulations. Probabilistic inversion approaches used here are based on Monte Carlo calculations which involve multiple parameters sets (samples of the parameter space) sorted in their *a priori* probability density function (PDF)  $\rho(\mathbf{m})$  and subsequent forward modeling using the physical model. This allows the identification of an *a posteriori* PDF  $\sigma(\mathbf{m})$  which describes in a more exhaustive way the distribution of the parameters yielding the best simulations. Among the Monte Carlo simulations which sample the parameter space, those simulations which minimize the misfit function samples in fact the *a posteriori* joint PDF  $\sigma(\mathbf{m})$ . From a theoretical standpoint, the posterior PDF is given by Tarantola (2005, chap. 2):

$$\sigma(\mathbf{m}) = L(\mathbf{m}) \times \rho(\mathbf{m}) \quad (8)$$

where  $L(\mathbf{m})$  is the likelihood function which writes:

$$L(\mathbf{m}) = k \times e^{-S(\mathbf{m})} \quad (9)$$

and  $k$  is a normative constant which ensures that the integral of  $\sigma(\mathbf{m})$  over the parameter space equals 1. Therefore, the *a posteriori* PDF represents an updated version of the *a priori* PDF based on an additional information provided by model-data comparison though

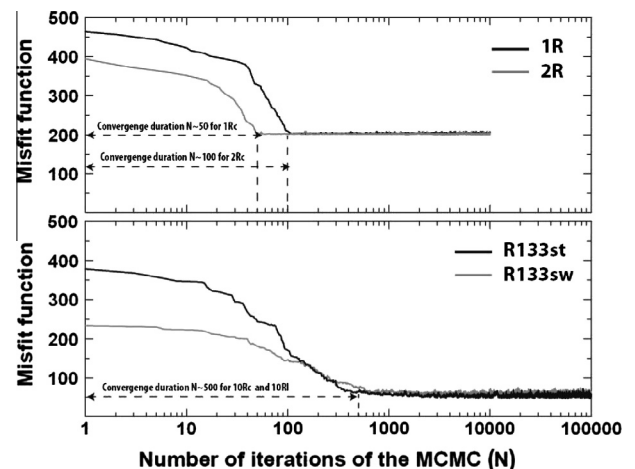
the likelihood function, whence the Bayesian approach. The sampling of the parameter space is thus crucial since it allows describing the joint PDF  $\sigma(\mathbf{m})$  and possibly the marginal PDF of all the model parameters  $\mathbf{m}(m_1, \dots, m_n)$ . The desirable identification of these parameters distributions is efficiently performed in the course of guided (towards maximum likelihood parameter space regions) random walk Monte Carlo inversion. This is also known as the Markov Chain Monte Carlo (MCMC) inversion approach. The popular MCMC inversion based on the Metropolis algorithm, briefly outlined below, that is adapted for *a priori* uniform distributions (Tarantola, 2005, chap. 2) was used in this study. Let's consider a current position  $\mathbf{m}_i$  of the random walk in the parameter space, and a potentially new position  $\mathbf{m}_j$  created by means of a random perturbation of  $\mathbf{m}_i$ . According to the Metropolis algorithm, the probability  $P$  to accept the displacement from  $m_i$  to  $m_j$  is:

$$P \begin{cases} 1 & \text{if } S(\mathbf{m}_i) > S(\mathbf{m}_j) \\ \frac{L(\mathbf{m}_j)}{L(\mathbf{m}_i)} = \exp(-\Delta S) & \text{if } S(\mathbf{m}_i) < S(\mathbf{m}_j) \end{cases} \quad (10)$$

where  $\exp(-\Delta S) = \exp(-S(\mathbf{m}_j) + S(\mathbf{m}_i))$ . Consequently, the perturbed model is accepted if it improves the data fit and can be accepted as well with a probability  $\exp(-\Delta S)$  if it increases the misfit. This latter occurrence allows leaving some local minima of the misfit function to explore all the likelihood maxima. From a practical stand point, a value is sorted in a uniform distribution between 0 and 1. If it is lower than  $\exp(-\Delta S)$  (which occurs with a probability  $\exp(-\Delta S)$ ) then the unfavorable displacement is accepted. From an initial position, the Metropolis algorithm outlined above is iteratively repeated  $N$  times. This probabilistic inversion algorithm converges towards the minima of the misfit function yielding an efficient sampling of the *a posteriori* PDF of the parameters.

## 6. Results

The MCMC inversion approach was used to ascertain the recharge values involved in the climatic scenario, in  $\text{mm yr}^{-1}$ , using *a priori* uniform distribution over the intervals [0,10] for  $\text{Rg}$ , the recharge rate during the glacial periods, [2;36] for  $\text{Rh}_1$ , and [0;100] for  $\text{Rh}[2, \dots, 9]$  the recharge rate during the interglacial period. The piston flow model assumes the same recharge scenario for the considered outcrops but considers different porosity for the



**Fig. 5.** Evolution of the misfit function as a function of the iteration number of the MCMC inversion for the four scenarios: (i) scenario with a unique recharge value (scenario 1R), (ii) square-wave scenario (scenario 2R) with the recharge rate during interglacial periods and glacial periods assumed constant, i.e., two parameters, one  $\text{Rg}$  and  $\text{Rh}$ , (iii) square-wave scenario (R133sw) with 9 humid recharge rate values and one glacial value and, (iv) saw-tooth scenario (R133st) with 9 humid recharge values and one glacial value.

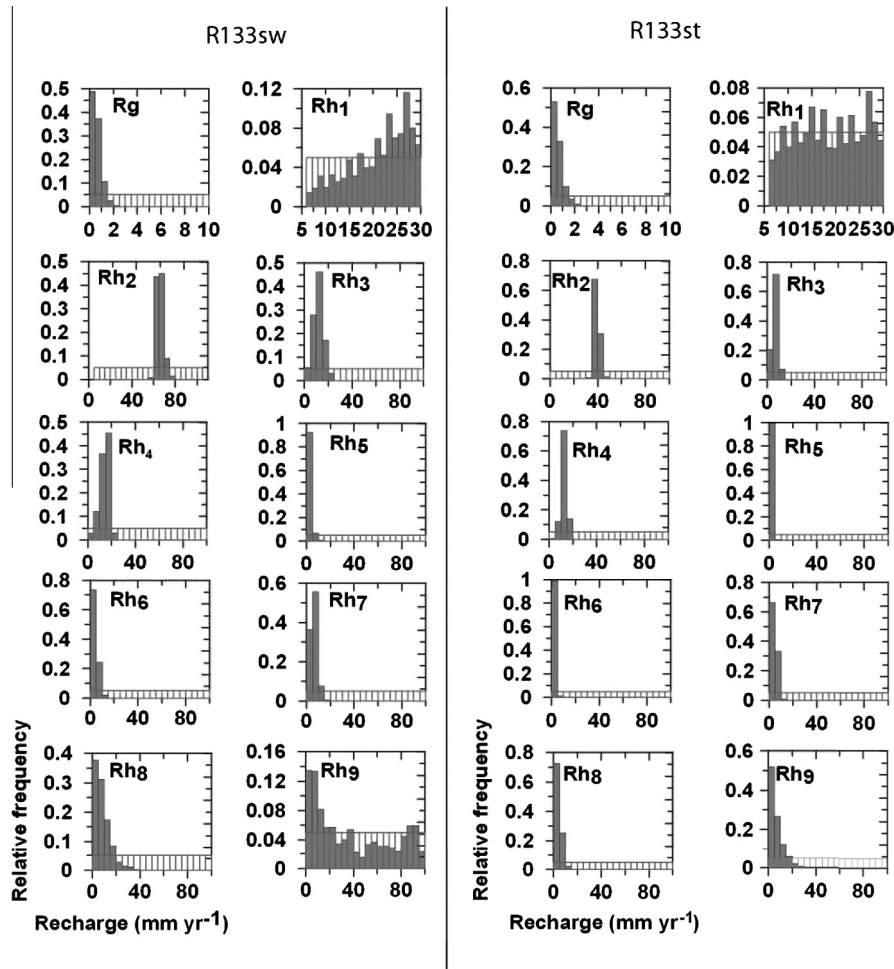


Fig. 6. Computed recharge rate values (in mm a<sup>−1</sup>) for the square-wave scenario (R133sw) and the saw-tooth scenario (R133st). *A priori* and *a posteriori* distributions of the recharge values from the sampling of the MCMC inversion using are shown in open gray bar and filled gray bar, respectively.

two formations. To assess the sensitivity of our overall approach to initial conditions, the MCMC inversion was carried out for four distinct recharge scenarios: (i) a scenario with a unique recharge value (scenario 1R), (ii) a square-wave scenario (scenario 2R) with the recharge rate during interglacial periods and glacial periods assumed constant, i.e., two parameters,  $R_g$  and  $R_h$ , (iii) a square-wave scenario (R133sw) with 9 humid recharge rate values and one glacial value and, (iv) a saw-tooth scenario (R133st) with 9 humid recharge values and a glacial value. Fig. 5 presents the value of the misfit function for these four scenarios as a function of the MCMC iteration number  $N$  using the Metropolis algorithm. As expected after the discussion in Section 5.1, the high misfit value

of the 1R simulation, about 200, illustrates how the constant 10 mm yr<sup>−1</sup> recharge value fails in reproducing the measured <sup>36</sup>Cl data. Although the 2R scenario does not improve the misfit value (also around 200, Fig. 5), it provides average recharge rates of 2 and 20 mm yr<sup>−1</sup> for the glacial and interglacial periods respectively. We used these estimates as *a priori* values, for the more complex MCMC inversions involving 10 parameters (R133sw, R133st). This step allows a quite fast convergence of the MCMC inversion process as illustrated in Fig. 5 where the misfit function

**Table 2**  
Calculated recharge rate (mm yr<sup>−1</sup>) for <sup>36</sup>R<sub>i</sub> = 133 × 10<sup>−15</sup> at at<sup>−1</sup> with the square-wave scenario (R133sw), with the saw-tooth scenario (R133st) and for a saw-tooth scenario considering <sup>36</sup>R<sub>i</sub> = 150 × 10<sup>−15</sup> at at<sup>−1</sup> (R150), <sup>36</sup>R<sub>i</sub> = 160 × 10<sup>−15</sup> at at<sup>−1</sup> (R160), <sup>36</sup>R<sub>i</sub> = 170 × 10<sup>−15</sup> at at<sup>−1</sup> (R170), <sup>36</sup>R<sub>i</sub> = 175 × 10<sup>−15</sup> at at<sup>−1</sup> (R175), <sup>36</sup>R<sub>i</sub> = 180 × 10<sup>−15</sup> at at<sup>−1</sup> (R180) and <sup>36</sup>R<sub>i</sub> = 200 × 10<sup>−15</sup> at at<sup>−1</sup> (R200).

| Rh | R133st | R133sw | R150 | R160 | R170 | R175 | R180 | R200 |
|----|--------|--------|------|------|------|------|------|------|
| 1  | 18.6   | 21.2   | 18.2 | 19.3 | 18.7 | 19.4 | 17.8 | 19.6 |
| 2  | 39.0   | 65.7   | 36.1 | 34.2 | 30.1 | 21.4 | 17.3 | 18.5 |
| 3  | 6.8    | 12.0   | 4.5  | 5.0  | 8.2  | 16.5 | 19.7 | 15.9 |
| 4  | 13.0   | 14.4   | 15.6 | 9.5  | 5.2  | 3.8  | 4.8  | 4.6  |
| 5  | 1.9    | 2.7    | 3.5  | 12.6 | 19.2 | 21.0 | 20.5 | 12.1 |
| 6  | 2.1    | 4.1    | 1.8  | 1.7  | 1.7  | 1.8  | 2.1  | 10.2 |
| 7  | 4.5    | 6.3    | 3.8  | 2.5  | 2.0  | 1.8  | 1.7  | 1.7  |
| 8  | 4.1    | 8.6    | 10.5 | 7.0  | 5.6  | 5.3  | 5.2  | 2.9  |
| 9  | 6.7    | 39.0   | 6.5  | 18.2 | 49.6 | 12.2 | 35.6 | 7.0  |
| Rg | 0.5    | 0.6    | 0.7  | 0.7  | 0.6  | 0.5  | 0.5  | 0.6  |

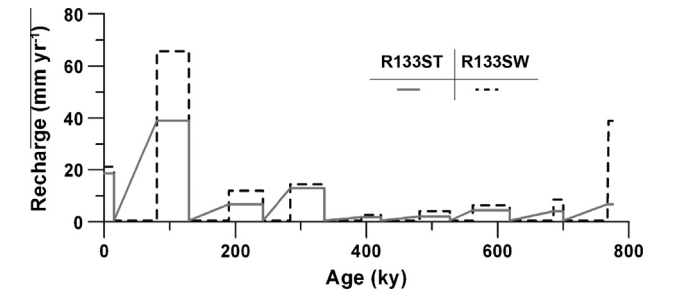
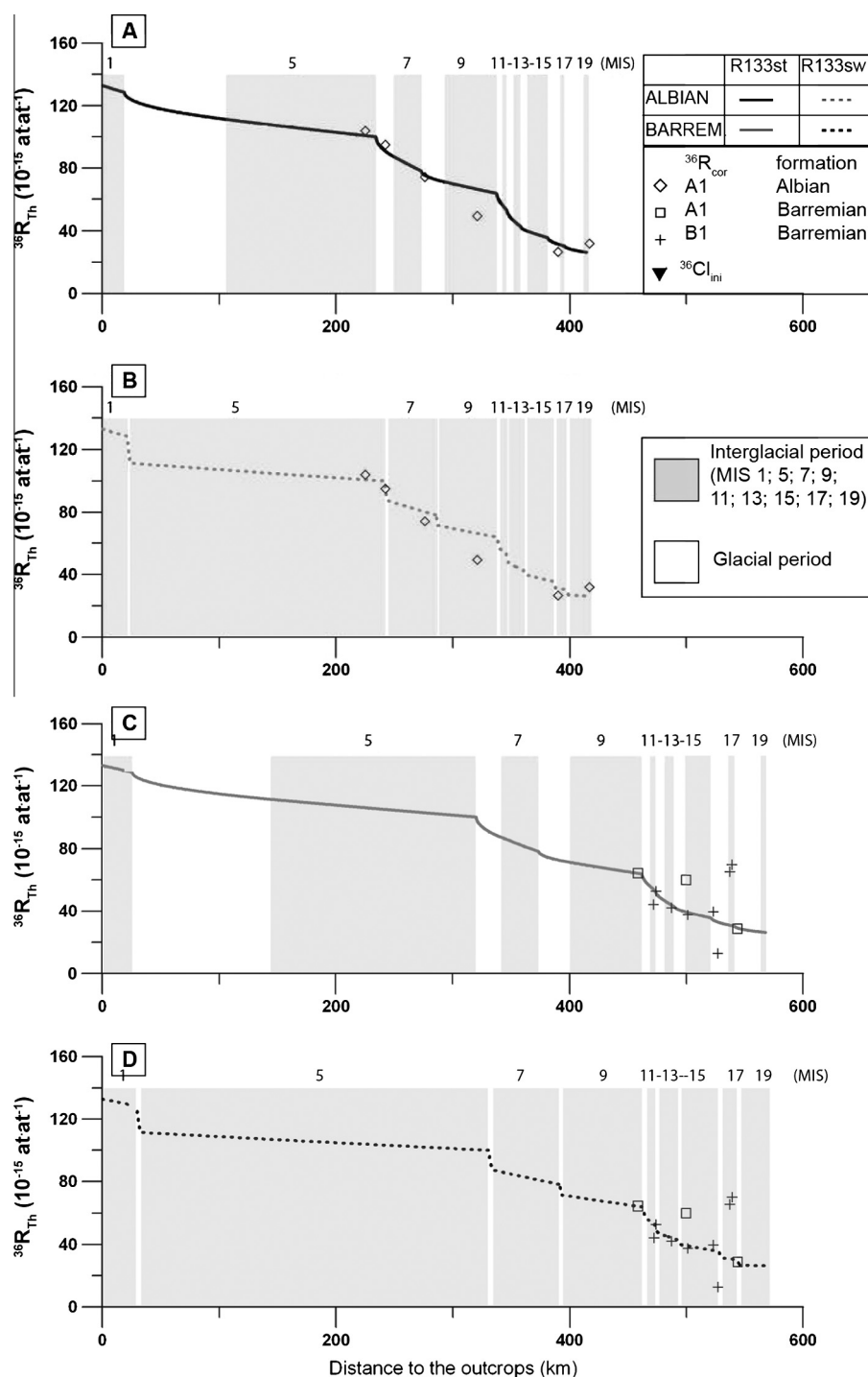


Fig. 7. Mean recharge rate obtained by MCMC sampling, for the R133st (gray line) and R133sw (dotted black line) scenarios.

reaches a plateau after about 100 iterations. During these 100 iterations, the MCMC method explores the parameter space positions of low likelihood (high misfit) before it converges towards the desired regions. The plateau corresponds to the sampling of the *a posteriori* joint distribution of parameters. The rigorous identification of the marginal distribution for all the parameters would require a rather complex mathematical treatment involving integral calculations of the joint distribution (Tarantola, 2005, chap. 2). But, according to Sambridge and Mosegaard (2002), for a large

sampling of the *a posteriori* distribution function, the marginal distributions can be simply identified by a statistical treatment of all the calculated values after the plateau, for each individual parameter. For the sake of simplicity, this approximate approach was used.

Fig. 6 shows the *a priori* (uniform) and the *a posteriori* distributions using histogram representations for the 10 parameters of scenario R133st and R133sw. Relatively well-constrained values (unimodal distributions) are obtained for all the recharge values



**Fig. 8.** Comparison between the  $^{36}\text{R}_{\text{cor}}$  data and the  $^{36}\text{R}_{\text{Th}}$  calculated using the recharge rates obtained from the MCMC inversion for the R133st and R133sw scenario. (A) The square-wave scenario for the Albian, (B) the saw-tooth scenario for Albian, (C) the square-wave scenario for Barremian and (D) the saw-tooth scenario for Barremian. Gray and white areas illustrate the distance covered during interglacial and glacial periods respectively (from results in Table 2 in Eq. (4)).

except that of the first humid period (Rh1) in both scenario and for the ninth (Rh9) humid periods of the square-wave scenario (R133sw). Although the use of the mean is inappropriate for the non-unimodal distributions, such mean values were calculated in every case for practical representation.

These recharge rates are represented in Fig. 7 for both R133st and R133sw scenarios (see also Table 2). For the two scenarios, a glacial recharge of  $1 \text{ mm yr}^{-1}$  or less is obtained while humid period recharge episodes range from a few  $\text{mm yr}^{-1}$  to 40 and  $65 \text{ mm yr}^{-1}$  for scenarios R133st and R133sw respectively. Recharge rate values obtained for the square-wave scenario (R133sw) are systematically higher than for the saw-tooth scenario due to the shape of the glacial period. For both scenarios, the maximum value is obtained for the second humid period (Fig. 7). Fig. 8 represents the comparison between  $^{36}\text{Cl}$  data ( $^{36}\text{R}_{\text{corr}}$ ) and  $^{36}\text{R}_{\text{Th}}$  simulated by the piston-flow model using alternatively the saw-tooth (R133st) and the square-wave (R133sw) scenarios with the mean values of recharge obtained from the MCMC inversions. This figure highlights the importance of the initial value. The latter has been defined according to hypothesis on measurements (see Section 4.2). In order to discuss these hypotheses, we apply sensitivity tests: we keep all the same parameters except the initial  $^{36}\text{Cl}/\text{Cl}$  and run new simulations using  $^{36}\text{R}_i = [150; 160; 170; 180; 200]$ , defining the R150, R160, R170, R180 and R200 scenarios respectively. Only the saw-tooth scenario was employed because this shape better describes the abrupt warming at the termination of a glacial period and a more gradual transition between the interglacial and the glacial period as described in Cheng et al. (2009), Wolff et al. (2009), and Barker et al. (2011). The mean of the recharge values are presented in Table 2. A best fit, i.e., misfit function values of 47; 43; 39; 38; 41 and 45 are obtained for R150, R160, R170, R175, R180 and R200 respectively, while 54 was obtained for the reference value  $^{36}\text{R}_i = 133 \times 10^{-15} \text{ at } \text{at}^{-1}$ . In the following part, these alternative initial values are discussed using independent climatic evidences.

## 7. Discussion

In order to put our reconstructions in perspective of paleoclimate data, we can use the recent compilation of Lang and Wolff (2011), who compared the relative intensity of glacial and interglacial periods over the nine climate cycles covering the last 800 kyr, based on available records of foraminiferal benthic and planktonic  $\delta^{18}\text{O}$ , sea surface temperature (SST), ice  $\delta D$  and  $\text{CH}_4$  and terrestrial proxies. We designed a crude common qualitative scale of strength of the interglacial periods by summing the individual rank of each interstadial in the 36 records used by Lang and Wolff (2011), for direct comparison with the rank of the recharge simulated in the corresponding period in our model, following the same assumption that stronger interglacial would have been associated with wetter conditions over North Africa (see Table 3). The results of this

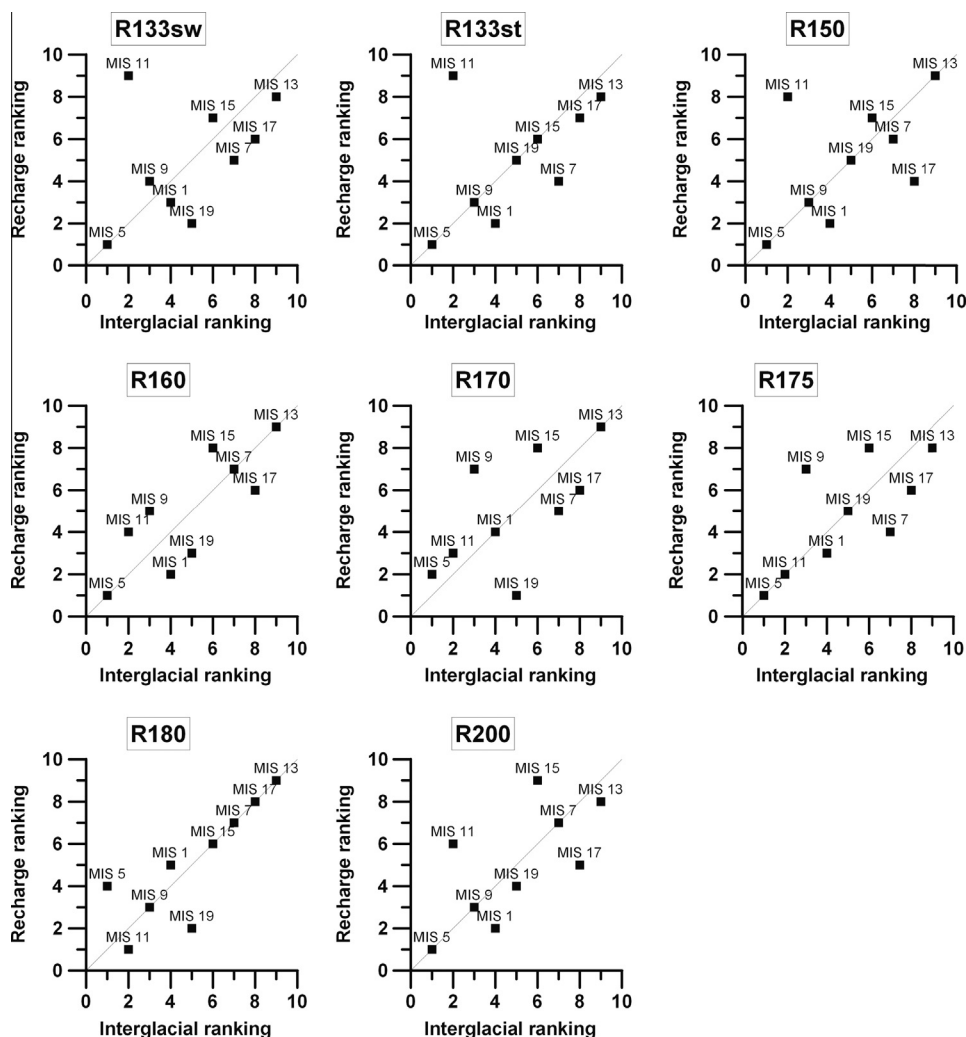
comparison are shown in Fig. 9, where is also illustrated the sensitivity of the respective ranking of the recharge intensities to the initial value  $^{36}\text{R}_i$  chosen for the simulations. It is then noticeable first that our model simulates relatively well the clear cut contrast between the most recent climate cycles (#1, 9, 5), that are both intense and humid, and the more ancient ones (#13, 15, 17), weaker and drier. This conclusion, which is well accepted in the climate record, is also robust in our hydrologic model, since it would not be possible to fit the observed data in any scenario where more water would have been introduced into the aquifer early instead of late during the period of interest.

A significant exception to this general agreement is the misfit for stage 11, which is known as one of the most prominent interglacial in the climate record, both in duration and in intensity, while trivial in our recharge reconstructions R133sw, R133st, and R150 (Fig. 9). Nevertheless, we observe that this misfit vanishes when higher values are used for the initial  $^{36}\text{R}_i$  composition (R170 to R180), underlining the weight of this parameter on the simulation results. This may reflect that our best-choice starting guess for this parameter may be under-evaluated or may have varied through time due to geomagnetic modulation of the  $^{36}\text{Cl}$  production (Baumgartner et al., 1998; Muscheler et al., 2005). Another interesting feature in Fig. 9 is that in all simulations but one, stage 5 is dominant in the series, and in particular, larger than the Holocene. In our model, this result comes from the narrow window of *a priori* values selected for Rh1 in the inversion calculation, based on existing paleoclimate reconstructions. This means that considering the plausible range of precipitation and infiltration values generally accepted for the Holocene, more water must have been recharged during previous stages, and preferentially during the last interglacial, in order to fit the spatial distribution of the  $^{36}\text{Cl}$  data. This is again coherent with marine (Osborne et al., 2008) or continental (Causse et al., 2003; Geyh and Thiedig, 2008) archive, since as mentioned above, MIS 5 was seemingly characterized by particularly intense precipitations (Paillou et al., 2009).

Besides these paleohydrologic inferences, another interesting aspect of our model is the demonstration that the two sets of data from the Albian and Barremian layers cannot be reconciled by using the same set of parameters, and must thus be considered as separate flow lines. Our results show that the difference can be accounted for by using a slightly larger porosity in the Albian than in the Barremian level. However, alternative explanations could be considered. In particular, the same geometry was adopted for the recharge area of the two layers. Any uncertainty on the length of the outcrop ( $L$ ) propagates directly on the calculated distance  $X(t)$ . The relative difference between the distances calculated for the two layers is thus within the *a priori* uncertainty affected to  $L$  (i.e. 30%). Other hypotheses, related to possible variations of the porosity or dimensions of the aquifers along the flowpath, would require additional observations and a

**Table 3**  
Ranking of the strength index of Lang and Wolff (2011) compared to the ranking of the recharge rate we calculated for the R133sw, the R133st, the R150, the R160, the R170, the R175, the R180 and the R200 scenarios.

| MIS/Rh | Strength<br>(Lang and Wolff, 2011) | R133st<br>(this study) | R133sw | R150 | R160 | R170 | R175 | R180 | R200 |
|--------|------------------------------------|------------------------|--------|------|------|------|------|------|------|
| 1/1    | 4                                  | 2                      | 3      | 2    | 2    | 4    | 3    | 5    | 2    |
| 5/2    | 1                                  | 1                      | 1      | 1    | 1    | 2    | 1    | 4    | 1    |
| 7/3    | 7                                  | 4                      | 5      | 6    | 7    | 5    | 4    | 7    | 7    |
| 9/4    | 3                                  | 3                      | 4      | 3    | 5    | 7    | 7    | 3    | 3    |
| 11/5   | 2                                  | 9                      | 9      | 8    | 4    | 3    | 2    | 1    | 6    |
| 13/6   | 9                                  | 8                      | 8      | 9    | 9    | 9    | 8    | 9    | 8    |
| 15/7   | 6                                  | 6                      | 7      | 7    | 8    | 8    | 8    | 6    | 9    |
| 17/8   | 8                                  | 7                      | 6      | 4    | 6    | 6    | 6    | 8    | 5    |
| 19/9   | 5                                  | 5                      | 2      | 5    | 3    | 1    | 5    | 2    | 4    |



**Fig. 9.** Comparison between “Interglacial ranking” following the compilation of Lang and Wolff (2011) and “Recharge ranking” determined using the different recharge scenarios tested during this work (Table 3).

more complete hydro-dynamical model to be tested. Along the same line of reasoning must also be mentioned that the Barremian data are significantly more scattered than the Albian, which may reflect short-term variations both in the initial  $^{36}\text{Cl}/\text{Cl}$  ratio and in the recharge rate, the smoother trend of the Albian data being either just fortuitous, or reflecting a more efficient mixing of such variations within the mixing zone upon recharge.

## 8. Conclusion

In this study the inversion of the recharge in semi-arid area and over various interglacial/glacial cycles scale i.e., 775 kyr was proposed by means of a combined approach involving  $^{36}\text{Cl}$  data and a simple hydrodynamic model (piston flow). MCMC inversion approach was successfully implemented in order to explore the uncertainties associated with the methodology. This study allows discussion of the global climatic scenario applying to the outcrop area in the Atlas Mountains as well as the significance of the  $^{36}\text{Cl}$  data distribution from these outcrops. Tests of the recharge scenarios point out that a constant recharge fails to reproduce the  $^{36}\text{Cl}$  data and a variation in the recharge improve their modeling. The best agreement between the climatic proxies (Lang and Wolff, 2011) and the  $^{36}\text{Cl}$  measurements was obtained for the simulation using  $^{36}\text{R}_i = 175 \times 10^{-15} \text{ at at}^{-1}$ . Plausible computed recharge

values range from few  $\text{mm yr}^{-1}$  to more than  $60 \text{ mm yr}^{-1}$  (see Table 2) for recharge during interglacial period and around  $1 \text{ mm yr}^{-1}$  or less for recharge during glacial periods. Such estimates could be used as *a priori* values in a more complex transient hydrogeological model of the CI.

However, because of the lack of  $^{36}\text{Cl}$  data close to the outcrops, the constraint of the most recent recharge rates (Rh1 and Rh2) relies on independent modern estimation and hydrological modeling (see Section 5.2). Further investigation of the residence time of groundwater in these strategic areas by means of geochemical tracers ( $^{36}\text{Cl}$ ,  $^{14}\text{C}$ ) would greatly improve our results. After all, these time-varying recharge values clearly illustrate the fact that hydraulic steady-state conditions are a crude hypothesis to interpret geochemical tracers in the NWSAS. Our results were obtained in the specific context of the Atlas Mountains but a similar method could be used in other aquifers provided that the piston flow model is nearly valid, that the climatic context is known and some  $^{36}\text{Cl}$  data (or other tracers) are available. Such approach may open new perspectives to the use of large aquifer systems as climatic archives by providing quantitative, and not only qualitative, assessment of paleorecharge rates. In the present case, we provide further evidence that climate condition in the Atlas Mountains, and very likely over the Saharan realm, were significantly wetter during MIS 5 than during the Holocene period.

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