



## Research papers

# Changes in groundwater reserves and radiocarbon and chloride content due to a wet period intercalated in an arid climate sequence in a large unconfined aquifer



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## ARTICLE INFO

## Article history:

Received 8 August 2017

Received in revised form 20 November 2017

Accepted 21 November 2017

Available online 22 November 2017

This manuscript was handled by Corrado Corradini, Editor-in-Chief, with the assistance of Bartolomé Andreo Navarro, Associate Editor

## Keywords:

Environmental tracer

Aquifer recharge

Radiocarbon

Chloride

Groundwater age

Arid zone

## ABSTRACT

The concentration of atmospheric tracers in groundwater samples collected from springs and deep wells is, in most cases, the result of a mixture of waters with a wide range of residence times in the ground. Such is the case of an unconfined aquifer recharged over all its surface area. Concentrations greatly differ from the homogeneous residence time case. Data interpretation relies on knowledge of the groundwater flow pattern. To study relatively large systems, the conservative ion chloride and the decaying radiocarbon (<sup>14</sup>C) are considered. Radiocarbon (<sup>14</sup>C) activity in groundwater, after correction to discount the non-biogenic contribution, is often taken as an indication of water age, while chloride can be used to quantify recharge. In both cases, the observed tracer content in groundwater is an average value over a wide range which is related to water renewal time in the ground. This is shown considering an unconfined aquifer recharged all over its area under arid conditions, in which a period of greater recharge happened some millennia ago. The mathematical solution is given. As the solution cannot be made general, to show and discuss the changes in water reserve and in chloride and radiocarbon concentration (apparent ages), two scenarios are worked out, which are loosely related to current conditions in Northern Chile. It is shown that tracer concentration and the estimated water age are not directly related to the time since recharge took place. The existence of a previous wetter-than-present period has an important and lasting effect on current aquifer water reserves and chloride concentration, although the effect on radiocarbon activity is less pronounced. Chloride concentrations are smaller than in current recharge and apparent <sup>14</sup>C ages do not coincide with the timing, duration and characteristics of the wet period, except in the case in which recharge before and after the wet period is negligible and dead aquifer reserves are non-significant. The use of chloride concentration in springs as a proxy of chloride concentration in recharge to estimate recharge from atmospheric deposition leads to recharge value larger than the real one and it approaches the wet period recharge. Drawing inferences about radiocarbon data and recharge by the chloride balance method has rarely been taken into account before. It is important to consider the variable aquifer groundwater reserve. Current recharge estimation can be improved by careful selection of groundwater samples, supported by tritium and radiocarbon measurements.

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## 1. Introduction

Aquifer recharge is a complex result of precipitation, climate conditions and soil characteristics. Complexity increases in arid areas where diffuse recharge may be almost nil, except after

occasional intense rainfall storms of decadal to centennial frequency and due to mountain block transfer. This recharge is able to sustain small perennial springs in large, low permeability formations containing long renewal time groundwater reserves in aquifers and aquitards, including slowly draining perched aquifers inside the generally thick unsaturated zone and the high relief areas.

At millennial scale, recharge conditions have often changed, following the sequence of arid and hyper-arid (dry) and not so arid

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(wet) periods, due to climate variability. Current springs and relatively high water tables may be a remnant of previous wetter-than-present periods, which dwindle slowly (Herrera and Custodio, 2014a,b). This dwindling may be unnoticed during a human life span but not historically when old documents, archeological data and chrono-stratigraphic records of paleo-wetland deposits data are available. They allow to know how spring flow has changed over time. The effect of previous wetter-than-present periods may be also reflected in chloride content in out-flowing groundwater, which is smaller than in current average recharge, and also in modified radiocarbon content in total dissolved inorganic carbon (TDIC).

The use of atmospheric chloride deposition is a well-known method to evaluate average long-term recharge (Eriksson and Khunakasen, 1969; Sukhija et al., 1988; Wood, 1999; Alcalá and Custodio, 2015), although corrections have to be applied when estimating recharge in sloping aquifers by using spring flow and water samples pumped from deep wells as a proxy of chloride content in recharge (Custodio and Jódar, 2016). However, in arid areas with memory from past wet events, observed groundwater chloride contents would yield too large current recharge rates.

The application of radiocarbon ( $^{14}\text{C}$ ) to determine recharge is also a well-known method (Clark and Fritz, 1997; Cook and Herczeg, 2000; Mazor, 2004; McCallum et al., 2017), especially in cases in which groundwater renewal time takes from centuries to millennia.  $^{14}\text{C}$  half-life is  $T = 5430$  years, equivalent to a decay constant of  $\lambda = \ln 2/T = 1.21 \cdot 10^{-4} \text{ yr}^{-1}$ . Since only a part of total inorganic dissolved carbon (TDIC) in groundwater may be of biogenic origin, or from the atmosphere in bare soils, corrections are needed to determine how much of the initial activity remains after some time (Fritz and Fontes, 1986; Mook, 2002; Han et al., 2012; Plummer and Glynn, 2013). The  $^{14}\text{C}$  activity of TDIC in the moment of generation  $A_0$  is calculated and this value is compared to the actual measured activity  $A$ , to give an age,  $t = (1/\lambda) \cdot \ln(A_0/A)$ . As the water sample is actually a mixture of waters with different transit times (ages), this is really an apparent age (Jones et al., 2006; Fiori and Russo, 2008; Maxwell et al., 2009; Engdahl and Maxwell, 2015).

In arid areas, where current recharge is considered nil, it is often assumed that the apparent water age is the time since a previous wet period. This is a risky assumption that has to be carefully discussed according to the aquifer conceptual flow model, taking into account that the sampled water is actually, in most cases, a mixture of a wide range of ages (transit times). Similar considerations were presented for perennial stream networks by McGuire and McDonnell (2006) and McDonnell et al., (2010).

In relatively large mountain range areas under arid conditions and with very low permeability, where groundwater flows through thick formations, a wetter-than-present period producing a large recharge is revealed through a noticeable groundwater reserve augmentation. In the case of exponential reserve depletion time of a few millennia, the increased groundwater reserves may last millennia and feed springs during a long arid period after the wet one.

This paper is a contribution to the understanding of the effect on groundwater reserve and chloride and radiocarbon content that a wetter-than-present period, with enhanced recharge, has on data collected from springs in arid lands, in the case of a water table (unconfined) aquifer. This is inspired in a mountain range case under hyper-arid conditions, where aquifer recharge is very sensitive to variable climate conditions during the past millennia. To quantify the results, a very simplified and generalized setting is worked out, in which recharge is homogeneously produced all over the aquifer surface and subjected to a millennial long wet period between two long dry periods. This setting allows obtaining an equation that can be solved with readily available numerical

methods. Real cases may differ from these simplifications, but results allow addressing the problem of uncertainty of corrected radiocarbon and chloride data in a hydrogeologically realistic framework.

As results cannot be made independent of recharge and aquifer characteristics, two general scenarios have been created, which are loosely based on actual studies being carried out in the hyper-arid desert of Northern Chile, and on authors' observations in the arid southern areas in the Canary Islands. However, this paper is not a case study, but an abstraction and generalization inspired in real cases. The objective is to get results that can be applied to a variety of circumstances.

The effect of variations in recharge rate along time on inferred water age has been investigated by Massoudieh (2013), Engdahl and Maxwell (2015) and Engdahl et al. (2016). Nevertheless, the related chloride and radiocarbon content changes in recharge for aquifers located in arid areas have rarely been considered before. This is the main subject of this work. A first approach was presented in Custodio and Custodio-Ayala (2014), although the solutions in this paper were only qualitative as the closed-form mathematical solution was not accurate enough all over the range values of interest.

## 2. Groundwater mixing in samples. Exponential flow aquifers

The mathematical treatment of the inflow-outflow of constant or time variable tracer concentration in an aquifer relies on simplified conceptual groundwater flow models using lumped parameters which apply diverse transfer functions to uniformly recharged aquifers (Zuber, 1986; Małoszewski and Zuber, 1982, 1996). A recent synthesis of the mathematical treatment and updated references can be found in Jódar et al. (2014). The relationship between groundwater age and age dating has been considered by Bethke and Johnson (2008). The case of linearly varying recharge in unconfined sloping aquifers is developed in Custodio and Jódar (2016).

Under non-dispersive and homogeneous conditions, for a confined aquifer in which recharge is only produced in a localized area up flow, or for in-transit recharge (percolation) through the unsaturated zone, the piston (plug) flow model with similar flow paths is a reasonable approach. This model is tacitly assumed when corrected apparent radiocarbon age is equated to groundwater age.

The piston flow does not represent the real behavior of groundwater in many common circumstances of high dispersion or distributed recharge over a large area. Water samples from regional springs or from deeply penetrating, long-screened wells are often a mixture of waters from different flow lines over a wide range or transit times.

The apparent groundwater age  $t$  [T] may greatly differ from the average renewal time of water in the aquifer  $\tau$  [T], defined as  $\tau = V/R$ , being  $V$  [L] the water depth in the system (reserve) and  $R$  [ $\text{LT}^{-1}$ ] the recharge.

In a pure piston-flow aquifer without geochemical effects on radiocarbon content, with no mixing during sampling,  $t$  will vary according to the sampling point, by increasing its value down gradient from the recharge zone along the flow path. Both,  $t$  and  $\tau$  will always yield the actual residence time at a point in the aquifer. When all flow lines have the same flow time, then  $t$  is equal to  $\tau$  at the aquifer's discharge point. Otherwise,  $\tau$  will only equal  $t$  for a flow line corresponding to the average residence time. Most discharge points would have either shorter or longer residence times than  $\tau$ . In a completely mixed aquifer, the actual residence time would be equal to  $\tau$  everywhere, but  $t$  would be smaller than  $\tau$  because mixing is linear and radioactive decay is nonlinear. This is not a consequence of mixing in the aquifer or during sampling,

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