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Comment on "Improving noble gas based paleoclimate reconstruction and groundwater dating using ^{20}Ne/^{22}Ne ratios," by F. Peeters et al. (2003) *Geochim. Cosmochim. Acta*, 67, 587–600

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1. INTRODUCTION

In their paper, Peeters et al. (2003) propose a new isotopic method for choosing an appropriate model for the interpretation of noble gas concentration in groundwater. They claim that a rigorous distinction between current available models is possible using ^{20}Ne/^{22}Ne ratios, and that this approach improves consistently noble gas based paleoclimate reconstruction and groundwater dating. This promising method is applied to a large sedimentary aquifer, the Continental Terminal in southwestern Niger.

Our comment focuses mainly on part 3 (study area and data) and part 4 (results and discussion) of the paper. Our main objective is to show that, even using ^{20}Ne/^{22}Ne analysis, an incomplete knowledge of the recharge process seriously biases noble gas interpretation. Consequently, in our opinion, some conclusions of the paper should at least be nuanced and reconsidered. A thorough understanding of the aquifer recharge process thus appears of the utmost importance when dealing with the interpretation of noble gas concentrations in groundwater.

Before discussion and without any link with the core of this comment, it must be noted that Eqn. 5 of the concerned paper is in fact a mix-up of two distinct equations (e.g., in Schlosser et al., 1989):

$$^{4}He_{ter} = {}^{4}He_m - {}^{4}He_{atm} \quad (5)$$

$$^{3}He_{tri} = {}^{3}He_m - {}^{3}He_{atm} - {}^{4}He_{ter} \cdot ({}^{3}He/{}^{4}He)_{ter} \quad (6)$$

where symbols are the same as in the discussed paper.

2. INDIRECT RECHARGE PROCESS

The hydrogeological context of the study area is well known and has been described in detail for the last decade. We believe that, owing to the natural complexity of the Continental Terminal (CT) aquifer, a more precise description is necessary for noble gas interpretation. For this study, the following points should be retained:

The CT is a large (150,000 km^2) and complex multilayered aquifer. Caution is therefore required for a representative geochemical sampling. For instance, natural leakages occur between the unconfined aquifer and confined aquifers at discrete places east of 3°E (Daddy Gaoh and Dassargues, 1995; Guéro et al., 2003). Recent investigations of the water table revealed that, following the regional land clearing observed for the past 50 yr, groundwater levels have risen by a few metres in most parts of the basin in response to a recharge increase of about one order of magnitude (Leduc et al., 2001; Favreau et al., 2002; Guéro et al., 2003). These spatio-temporal changes often require both hydrodynamic and geochemical data to be identified, and must be taken into account for selecting an adequate data set. Because the unconfined aquifer cannot be considered as a stable system, care should be taken when interpreting tracers in terms of groundwater dating.

Most of the groundwater recharge is indirect. During the rainy season from June to September (90% of the annual rainfall), intense rainfall events of convective origin (half of the annual rains falling at rates > 35 mm h^{-1}; Lebel et al., 1997) produce Hortonian runoff that rapidly (within 1–3 h) concentrates in temporary pools, natural outlets of endoreic basins of few square kilometres. All hydrological data indicate that most of the aquifer recharge occurs under the pools (Desconnets et al., 1997; Martin-Rosales and Leduc, 2003). Volumes infiltrated from a pool to the aquifer are frequently 10,000 m^3 yr^{-1} but can reach 100,000 m^3 yr^{-1} in some places or during the rainiest years. In most cases, infiltration takes place over a few hours or days. Consequently, as illustrated in Figure 1, transient water table mounds, up to a few metres of amplitude, are created below infiltrating pools (Leduc et al., 1997; Favreau et al., 2002). From a geochemical point of view, this indirect recharge process is confirmed both by (1) an absence of isotopic (δ^2H, δ^{18}O) fractionation of groundwater due to evaporation (Favreau et al., 2002) despite a typical semiarid environment and by (2) abrupt chemical changes in groundwater associated with recharge events near infiltrating pools (Elbaz-Poulichet et al., 2002). Elsewhere in the landscape, there is no infiltration deeper than 4 m below the soil surface (Peugeot et al., 1997), with the only possible exceptions of narrow banded vegetation on the plateaux (Galle et al., 1999) and of gullies on the sandy slopes (Esteves and Lapetite, 2003).

The Dallols, located on Figure 2 of the discussed paper, represent an exception where direct recharge may occur due to (1) lower slopes, (2) coarser quaternary sediments and (3) shallow groundwater table. However, they represent only 10% of the study area and most of the discussed groundwater sampling took place outside these large valleys.

Brennwald et al. (2001) are cited to advocate for a time-lag

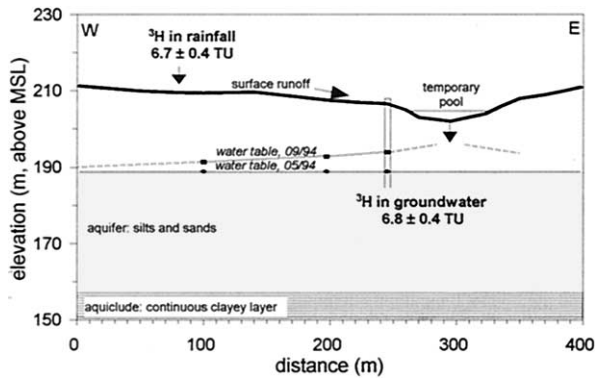* Author to whom correspondence should be addressed (guillaume.favreau@msem.univ-montp2.fr).

Fig. 1. Cross section (west to east) at Wankama (60 km east of Niamey) showing the transient potentiometric mound of the water table below the temporary pool before (May 1994) and after (September 1994) the rainy season. The ^{3}H content in groundwater (February 1995) at a distance of 50 m from the pool and in rainfall in Niamey (September 1994) are also displayed.

in ^{3}H input to groundwater due to transport through the CT unsaturated zone. This study is based on a theoretical model of gaseous transport (Cook and Solomon, 1995) that clearly refers to diffuse groundwater recharge as the main process. This process is poorly significant in southwestern Niger: hydrodynamic measurements as well as geochemical investigations conducted for more than a decade throughout the whole area, both referring to surface and ground waters, have proved that the effective groundwater recharge process is mainly indirect, occurring only where surface runoff concentrates (Fig. 1).

3. IMPLICATION FOR NOBLE GAS INTERPRETATION

A first implication of a rapid, massive indirect recharge process is the absence of any time-lag for ^{3}H input to the water table. Postbomb ^{3}H values between 5 and 26 TU were measured in several parts of the unconfined aquifer during the 1990s, at places where the groundwater table lies between 20 and 40 m below the soil surface (Leduc et al., 1996; Guéro et al., 2003). These ^{3}H contents could be explained by (high) diffuse recharge, but because radioactive decay would reduce ^{3}H content during the water transport through the large unsaturated zone of the CT aquifer, these postbomb ^{3}H values are better explained as a consequence of indirect recharge process. A definitive evidence for the absence of any time-lag for ^{3}H input to the saturated aquifer is reported for a local scale study (Fig. 1). At this site, for an infiltrating surface area near 30,000 m^2, groundwater recharge was ~75,000 m^3 in 1994 and the highest recorded amplitude of the water table mound was 5.1 m (September 1994). Considering these characteristics, local recharge was as high as 2500 mm yr^{-1} below this temporary pool. During the subsequent dry season the ^{3}H content was logically identical (6.8 TU, February 1995), within the analytical accuracy, to the ^{3}H content in 1994's rainfall (6.7 TU, September 1994). These results have important implications when applying the ^{3}H-^{3}He method, particularly for a reliable estimate of the tritiogenic ^{3}He.

Peeters et al. (2003) apply, among other approaches, the ^{3}H-^{3}He dating method to compare results of two gas exchange

models, the closed system equilibration model or CE-model, and the partial re-equilibration model or PR-model (these models are described in detail in part 2 of their paper). Using measured noble gas elemental concentrations and ^{3}He/^{4}He ratios as a data set, the CE-model indicates ^{3}He$_{tri}$ = 0.2 ± 0.3 TU for CT groundwaters of prebomb origin, whereas for the same samples, the PR-model indicates ^{3}He$_{tri}$ = 11 ± 1 TU. They conclude that the CE-model is more realistic, as they expect prebomb ^{3}H in groundwater to be close to zero as a consequence of radioactive decay during diffuse recharge through the unsaturated zone. However, because of instantaneous transfer of modern ^{3}H to the water table (Fig. 1), this conclusion has to be rejected. In fact, both model results differ significantly from a prebomb ^{3}He$_{tri}$ value that should be in the range of 3 to 6 TU in the northern hemispheric, continental West Africa (Leduc et al., 1996; Le Gal La Salle et al., 2001; Favreau et al., 2002), in the absence of any subsequent ^{3}He$_{tri}$ loss. An additional test included ^{20}Ne/^{22}Ne ratios in the data set. The predicted ^{3}He$_{tri}$ values of the CE-model did not change, whereas the new PR-model output was ^{3}He$_{tri}$ = 3 ± 1 TU. Although this result was disregarded by the authors, it appears more realistic in a context of indirect recharge. Clearly, because of the absence of any time-lag for ^{3}H input into groundwater, a consistency test of the ^{3}H-^{3}He water ages based on the comparison of ^{3}H$_{rech}$ with the historic ^{3}H concentration at recharge (as, e.g., Aeschbach-Hertig et al., 1998) is both practicable and needed to assess which model, using ^{20}Ne/^{22}Ne ratios, is the more reliable.

Another remark about interpretation of ^{3}H-^{3}He results concerns groundwater mixing from different aquifers. As outlined by Schlosser et al. (1989), mixing of waters with different ^{3}H-^{3}He values results in a non-linear behaviour of the ^{3}H-^{3}He age. However, in the data set of Figure 5 at least one groundwater sample is obviously the result of leakage between the CT2 and CT3 aquifers (number 30, data in Beyerle et al., 2003). At this sampling site, postbomb ^{3}H is measured in wells tapping the CT3 aquifer (12.3 ± 0.5 TU) whereas regional CT2 groundwaters are ^{3}H-free (Guéro et al., 2003); locally higher potentiometric levels in the CT3 than in the CT2 aquifer provide an explanation for the observed leakage. Consequently, ^{3}H-^{3}He results of this sample cannot be interpreted in term of groundwater age. Careful examination of the sampling sites and of local hydrodynamic conditions are a basic prerequisite when dealing with geochemical interpretation of groundwater.

A second implication of rapid infiltration of surface water to the water table is temperature of recharge. In the study area like in most parts of the Sahel, rainfall events induce a decrease in air temperature by up to 5 to 10°C, as a consequence of high rates of convective rainfall and of enhanced exchange in the lower atmosphere far from being saturated with water vapor (Taupin et al., 2000). As runoff events last only a few hours (Desconnets et al., 1997), surface water does not always reach thermic equilibrium with the soil. For instance, localized temperature measurements of runoff showed values in the range of 20 to 25°C in gullies, and in the range of 24 to 28°C in temporary pools the days after rain events (Desconnets et al., 1995). Because infiltration is a massive (typically several hundred to thousand millimetres per year below the pools) and quick process (a few hours to a few days; Desconnets et al., 1997) water reaches the water table below infiltrating pools at

a significantly lower temperature than the annual mean soil temperature of 32°C. Since 1998, groundwater temperature of the upper metres of the saturated aquifer recorded at 50 m from the infiltrating pool reported in Figure 1 have shown consistent values between 28 and 29°C throughout the year; similar temperatures for the upper metres of the unconfined aquifer were recently recorded in pumped wells near other infiltrating pools. In their paper, Peeters et al. (2003) consider the mean soil temperature of 32°C in the study area as a reference for recharge temperature. As the initial condition for noble gas in groundwater is set at the interface between saturated and unsaturated zone (Stute and Schlosser, 2000), considering the mean soil temperature of 32°C gives an overestimation of the real groundwater recharge temperature of $\sim$3°C. It is assumed that subsequent warming of groundwater within the saturated aquifer may occur (measured groundwater temperature in the range of 28 to 32°C; Le Gal La Salle et al., 2001; Favreau et al., 2002). However, the initial noble gas conditions remain determined by the characteristics of groundwater recharge below the pools.

4. CONCLUSION

Peeters et al. (2003) recognize that "it is still unclear which environmental conditions lead to the noble gas pattern" described by the available models in the literature. They conclude that this can be judged only from data on concentrations of noble gases and their isotopes. However, as for groundwater recharge in semiarid areas, "realistic estimation depends on first identifying flow mechanisms" (Lerner et al., 1990). As shown in this discussion, considering in southwestern Niger a theoretical diffuse recharge instead of the effective indirect recharge may lead to incorrect conclusions, both for (1) ^{3}H-^{3}He dating and (2) estimate of recharge temperature. Whatever the complexity of the geochemical model, it cannot bring relevant results if it is not based on a relevant schematization of the natural complexity. This implies that the main hydrological processes are well defined. In semiarid areas, this knowledge often requires mutual confirmation of geochemical and hydrodynamic informations. Further research is obviously needed to better link environmental conditions with noble gas pattern.

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