Author's copy, original version published in Geochim. Cosmochim. Acta, 2004

Pergamon

Geochimica et Cosmochimica Acta, Vol. 68, No. 6, pp. 1437–1438, 2004
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0016-7037/04 $30.00 + .00

doi:10.1016/j.gca.2003.08.013

Response to the comment by G. Favreau, A. Guero, and J. Seidel on "Improving noble gas based paleoclimate reconstruction and groundwater dating using $^{20}Ne/^{22}Ne$ ratios" (2003) *Geochim. Cosmochim. Acta*, 67, 587–600

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(*Received August* 1, 2003; *accepted in revised form August* 13, 2003)

1. INTRODUCTION

In groundwaters excess air and its fractionation is a common difficulty in the interpretation of noble gas concentrations with respect to recharge temperature but also with respect to the contribution of atmospheric He and thus to residence times based on the ^3H-^3He method. In our paper (Peeters et al., 2003) we demonstrated that data sets including Ne, Ar, Kr and Xe may not be sufficient to distinguish between different gas partitioning models. Including data on isotope concentrations and in particular including measurements of ^{20}Ne and ^{22}Ne provides additional constraints on the models and thus increases the sensitivity of the analysis to model choice. We demonstrated that in case of the CT-aquifers in Niger only the closed-system equilibration-model (CE-model, Aeschbach-Hertig et al., 2000) is compatible with the entire data set.

According to our analysis the samples from the CT3 considered in our paper (Peeters et al., 2003) have noble gas temperatures (NGT) ranging between 29.5 and 33 °C indicating that recharge in the quasi saturated zone at the soil air–ground water interface occurred at ~32°C (see also Beyerle et al., 2003). In addition we calculated the atmospheric contribution of ^3He and ^4He to the overall ^3He and ^4He concentration based on the heavier noble gas concentrations and the Ne isotopes. Assuming a constant terrigenic $^3He/^4He$ ratio then allowed us to determine the tritiogenic component of ^3He, $^3He_{tri}$, in our samples. Considering only samples without tritium we estimated prebomb $^3He_{tri}$ to be < 0.6 TU. For the purpose of illustrating the importance of determining the correct excess air model, we showed how the estimates of NGTs, ^3H-^3He ages, and prebomb $^3He_{tri}$ would change if other models describing excess air formation were used. However, the choice of the model cannot be based on any preference with regard to these secondary quantities, but must be derived from the noble gas data themselves. The central point of our paper is to show that a unique model choice is possible if Ne isotopes are included in the analysis.

The comment of Favreau et al. concentrates on the question whether recharge in Niger occurs locally at temporary pools immediately after strong rainfall events or infiltration takes place in a more spread out fashion over the entire region. This is apparently a hotly debated topic (Leduc et al., 2001; Bromley et al., 2002; Favreau et al., 2002b), but has not been addressed or discussed in our paper. We are therefore surprised by the comment which actually is not related to the central part of our paper but apparently was caused by a one-sentence side argument related to the low value of prebomb tritium and the cited abstract of Brennwald et al. (2001). Before providing a detailed response to the arguments by Favreau et al. we must again clearly state that our analysis of the noble gas data is firm and its outcome cannot be modified to satisfy hypotheses on the recharge mechanism in Niger proposed by Favreau et al. In contrast the data and the derived properties should be the basis to test hypotheses on recharge mechanisms.

2. PREBOMB ^3H AND TIME-LAG IN THE ^3H INPUT

Our estimate of prebomb tritium in the groundwater (0.2 ± 0.3 TU) follows directly from the calculated $^3He_{tri}$ concentration in tritium free groundwater and does not require assumptions on recharge mechanisms. Prebomb tritium in groundwater is expected to be lower than the poorly-known prebomb tritium content of precipitation because of ^3H decay during transport through the unsaturated zone. We consider our rather low value of prebomb tritium to be consistent with slow diffusive and advective transport through the unsaturated zone, as modeled by Brennwald et al. (2001). This model considered diffusive and advective transport in the unsaturated zone but did not include local recharge by preferential flow. According to the comment by Favreau et al. recharge occurs predominantly as a very fast process at temporary pools. Therefore, no reduction of tritium by a time-lag between precipitation and arrival at the water table would be expected. Favreau et al. question our prebomb $^3He_{tri}$ value by stating that it "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., 2002a)." These references contain no original data on the prebomb ^3H of the local precipitation. Favreau et al. (2002b) simply state: "The background level at the beginning of the 1950's is assumed to be 5 TU" citing Kaufmann and Libby (1954). We consider this assumption as highly uncertain, because the prebomb tritium data discussed by Kaufmann and

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Libby (1954) are from midlatitude sites (except one low (0.6 TU) value from Hawaii, a tropical oceanic site). Moreover, since Favreau et al. state that recharge has strongly increased in the past decades, predominant recharge pathways may have changed since the formation of the prebomb water. Assuming that prebomb ^{3}H of the local precipitation was indeed 3 to 6 TU, the small prebomb ^{3}H derived from the noble gas data suggests that in prebomb times a significant amount of ^{3}H must have decayed during transport through the unsaturated zone.

Present recharge mechanisms may be studied using tracer data of recently infiltrated water. The work by Brennwald et al. (2001) was motivated by significant differences between water ages based on CFC-11 data and the ^{3}H-^{3}He method suggesting that transport in the unsaturated zone delays CFC-11 concentrations. In their comment Favreau et al. suppose infiltration to be a very fast and localized process. In this case however one would expect CFC-11 and ^{3}H-^{3}He water ages to agree well at least locally near the pools. Since the two tracer ages disagree in our samples they apparently were not significantly affected by direct infiltration from temporary pools.

3. NOBLE GAS TEMPERATURES

According to the comment by Favreau et al. water infiltrating from temporary pools must have temperatures significantly below 32°C. Favreau et al. base their argument on the temperature of precipitation and on groundwater temperatures below 29°C measured near a temporary pool. However, the noble gas concentrations in our samples from the CT3 indicate that our samples have equilibrated with soil air at temperatures around 32°C. Hence the infiltration process suggested by Favreau et al. is not compatible with our data. There may be several possible explanations: (1) The water at the sites sampled by us did not infiltrate by the pool process proposed by Favreau et al. (2) The water infiltrated at the pools but had sufficient time to reach thermal equilibrium with the soil before it reached the water table (3) The water re-equilibrated after the fast infiltration process with soil air at ~32°C. (4) The temperature conditions at the single temporary pool investigated by Favreau et al. are not representative for the temperature conditions at temporary pools in the area investigated by us.

Our paper did not address such questions on recharge mechanisms in Niger. Its intention is to provide a rigorous method to analyse noble gas data which can then be employed for the calculation of NGTs and for ^{3}H-^{3}He dating. These techniques could be applied to resolve some of the open questions related to the recharge mechanisms in Niger. In particular ^{3}H-^{3}He dating near the temporary pools may help to constrain the travel times through the unsaturated zone and the vertical exchange within the CT3. Dating based on ^{3}H alone may not be very conclusive.

4. CONCLUSION

Since the publication of the paper by Peeters et al. (2003) some progress has been made concerning the processes leading to excess air formation and the fractionation of noble gas composition by laboratory experiments (Holocher et al., 2002, 2003; Kipfer et al., 2002). Nevertheless in field samples the best way to distinguish between the different gas partitioning models are isotope measurements, particularly measurements of Ne isotopes. The advantage of the noble gas analysis is that it does not require a recharge model. Neither NGTs nor ^{3}H-^{3}He ages depend on assumed flow mechanisms. The intention of measuring tracer data is to provide a broad data base to improve understanding of flow mechanisms. The hypotheses on recharge processes have to be tested against the data.

Acknowledgments—The Swiss Federal Institute of Environmental Science and Technology (EAWAG) and the Swiss National Science Foundation supported our research (SNF grants: #2000-061498.00 and 20-68191.02).

Associate editor: B. Marty

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