



ELSEVIER

SCIENCE @ DIRECT®

Earth and Planetary Science Letters 240 (2005) 827–829

EPSL

www.elsevier.com/locate/epsl

Discussion

A comment on “Helium sources in passive margin aquifers—new evidence for a significant mantle ^3He source in aquifers with unexpectedly low in situ $^3\text{He}/^4\text{He}$ production” by M. C. Castro [Earth Planet. Sci. Lett. 222 (2004) 897–913]

W. Aeschbach-Hertig*

Institute of Environmental Physics, University of Heidelberg, Germany Im Neuenheimer Feld 229 D-69120 Heidelberg, Germany

Received 7 June 2005; received in revised form 22 September 2005; accepted 22 September 2005

Available online 28 October 2005

Editor: K. Farley

In a recent article, M.C. Castro [1] uses a numerical groundwater model to study the transport of He isotopes in the Carrizo aquifer, Texas. The main result is that an unexpectedly large mantle ^3He component is present in this system, masked by the in situ produced He with a very low $^3\text{He}/^4\text{He}$ ratio. Suggested implications are that groundwater modeling is needed to fully understand the mixing and transport of He components of different origin and that the flux of mantle ^3He through the crust may previously have been underestimated.

I will show in the following that the results obtained in this study are hardly surprising and essentially could have been obtained by simple mixing calculations. I conclude that complex models are not needed to assess the mantle ^3He fraction in groundwaters, and that there is no need to re-evaluate any previous mantle He flux estimates. I start with a discussion of the mixing of two He components, as this calculation seems to have been misinterpreted.

1. He isotope mixing calculations

I consider the He isotope ratio of a sample (s) consisting of a mixture of a crustal (c) and a mantle

(m) He component, which leads to a simplified form of Eq. (1) of Castro [1]:

$$R_s = \frac{^3\text{He}_s}{^4\text{He}_s} = \frac{^3\text{He}_c + ^3\text{He}_m}{^4\text{He}_c + ^4\text{He}_m}. \quad (1)$$

This can be rewritten as:

$$R_s = \frac{^4\text{He}_c R_c + ^4\text{He}_m R_m}{^4\text{He}_s} = x R_m + (1 - x) R_c. \quad (2)$$

where R_c and R_m denote the $^3\text{He}/^4\text{He}$ ratios of the two components and x is the mantle ^4He fraction ($x = ^4\text{He}_m / ^4\text{He}_s$) in the sample. Solving Eq. (2) for x yields:

$$x = \frac{R_s - R_c}{R_m - R_c}. \quad (3)$$

Note that x is the mantle fraction of ^4He (which is essentially identical to the total He), but not of ^3He . The mantle fraction of ^3He is given by:

$$y = \frac{^3\text{He}_m}{^3\text{He}_s} = \frac{^4\text{He}_m R_m}{^4\text{He}_s R_s} = x \frac{R_m}{R_s}. \quad (4)$$

Castro [1] clearly got this wrong when she wrote that the He isotope ratio of the external He flux derived for the Carrizo aquifer of $R_{\text{ext}} = 1.9 \cdot 10^{-7}$ ($R_{\text{ext}}/R_a = 0.14$, where R_a is the atmospheric $^3\text{He}/^4\text{He}$ ratio) would indicate 1.5% mantle ^3He contribution if typical

* Corresponding author. Fax: +49 6221 54 64 05.

E-mail address: aeschbach@iup.uni-heidelberg.de.

mantle and crustal isotope ratios of $R_m = 1.2 \cdot 10^{-5}$ and $R_c = 2 \cdot 10^{-8}$ are assumed (p. 906 in [1]). In fact, these values yield a ^4He mantle fraction (x) of 1.5%, but a ^3He mantle fraction (y) of 90%. The same mistake is repeated on p. 910 in [1], where a R_{ext} value of $4 R_a$ is said to indicate a 46% mantle ^3He fraction, when in reality it is 46% mantle ^4He and 99.8% mantle ^3He . These results are indeed surprising at first sight, but they are a natural consequence of the extreme differences in the He isotope ratios of the different geochemical reservoirs. For illustration, consider the limiting case $R_c = 0$. In this case, there is no crustal ^3He at all, thus ^3He is always 100% of mantle origin, while the fractions for ^4He (and total He) remain nearly unchanged. Anyone familiar with these mixing calculations should therefore not be surprised that even a rather low $^3\text{He}/^4\text{He}$ ratio can indicate a high mantle contribution for ^3He , while the mantle fraction of ^4He is small.

A major issue in the paper of Castro [1] is the masking of a relatively high $^3\text{He}/^4\text{He}$ ratio of the external He flux to the aquifer by the in situ produced He with a very low $^3\text{He}/^4\text{He}$ ratio. This effect can entirely be understood in terms of simple mixing calculations, without a numerical model. For instance, the “quasi-instantaneous lowering of the $R_{\text{ext}}/R_a \dots$ as external helium enters the aquifer \dots from 0.14 to 0.055 at the base of the Carrizo” (p. 909 in [1]) is simply the consequence of two-component mixing. Using Eqs. (3) and (4) above, but inserting as endmembers the external flux with $R_{\text{ext}} = 0.14 R_a$ (instead of R_m), the in situ production with $R_{\text{in}} = 0.0025 R_a$ (instead of R_c), and the measured excess ratio R_{exc} of $0.055 R_a$ (for R_s) yields a relative contribution of the external flux of 38% to ^4He (x) and 97% to ^3He (y). These results agree very well with the model results shown in Fig. 6 in [1], but are derived from a simple mixing calculation.

Also the finding that even a roughly 6 times higher isotope ratio for the in situ production can explain the depression of the excess He isotope ratio discussed on p. 911 of [1] is fully explained by mixing: Increasing R_{in} to $0.0144 R_a$ (based on the ^3He production rates given on p. 911 and in Table 2 of [1]) and keeping $x = 0.38$, Eq. (2) with the above substitutions yields $R_{\text{exc}} = 0.062 R_a$, i.e., just slightly higher than measured, as found with the numerical model. Similarly, the strong reduction of hypothetical much higher values for R_{ext} at the base of the aquifer can simply be understood by the above mixing equations and fractions (e.g., according to Eq. (2) mixing 38% of external flux He with $R_{\text{ext}} = 1.4 R_a$ with 62% of in situ produced He with $R_{\text{in}} = 0.0025 R_a$ reduces the excess isotope ratio to 0.53

R_a , or by factor of 2.6 compared to R_{ext} , similar to the factor of 2.8 stated on p. 910 in [1]).

In fact, there is no reason why the numerical transport model should reveal anything about the change of the He isotope ratio that could not simply be explained by mixing of the different He components. The only mechanism by which the model could introduce an isotope fractionation that is not captured by the mixing calculations would be the different molecular diffusivities of ^3He and ^4He . But, as stated in the paper (p. 904 in [1]), the effects of this difference are negligible, since advection is dominant.

2. The use of He and ^{14}C data

The only piece of information the numerical model does provide, which could not have been derived from the He data alone, is the isotope ratio of the external flux, or, equivalently, the mixing ratio of the external and in situ produced components. There is however a way how this information could have been derived directly from data. If we knew the age (residence time) of the groundwater in the Carrizo, the in situ component could be calculated from the production rates. In fact, there are ^{14}C ages for the Carrizo aquifer, used in an earlier paper of Castro et al. [2] and in the paper of Stute et al. [3], where the He data originate from. Unfortunately, of the samples with He data listed in Table 1 of [1], only very few have a ^{14}C age listed in Table 1a of [2]. Looking for a relatively old sample, where ^{14}C ages and ^4He excesses should have comparatively small relative errors, I used sample TX04 to demonstrate how the isotope ratio of the external flux can be estimated from original data. From the ^{14}C age of this sample of 32.7 kyr [2], and the ^4He production rate of $3.3 \cdot 10^{-18} \text{ mol m}^{-3} \text{ s}^{-1}$ (Table 2 in [1]), one can calculate that $3.4 \cdot 10^{-6} \text{ mol m}^{-3}$ of ^4He have been produced in situ. This is only 20% of the total He excess of $16.8 \cdot 10^{-6} \text{ mol m}^{-3}$ at this site (Table 1 in [1]), thus 80% of the excess originate from the external flux. Using these fractions ($x = 0.8$) and $R_{\text{in}} = 0.0025 R_a$, the mixing equations tell us that the isotope ratio of the external flux R_{ext} has to be about $0.09 R_a$, in order to produce the observed excess isotope ratio R_{exc} of $0.07 R_a$ in this sample (or about $0.07 R_a$ to produce the typical R_{exc} of $0.055 R_a$).

This result differs significantly from that of Castro [1], who postulates that about 60% of the non-atmospheric He excess originate from in situ production and 40% from the external flux (see Fig. 6b in [1] for high distances with negligible atmospheric contribution) and consequently requires a higher R_{ext} of $0.14 R_a$ to ex-

plain the mean observed R_{exc} of $0.055 R_a$. The higher fraction of in situ produced He implies a higher age of the water (roughly by a factor of 3 for TX04). Thus, it appears to me that the numerical model calibrated with ^4He data is not consistent with ^{14}C data from the aquifer. This problem is even more obvious from a previous paper by Castro and Goblet [4], which apparently is based on the same model. The contradiction with the ^{14}C ages was resolved in that paper by simply discarding the ^{14}C ages and using model-derived age estimates instead. The authors of that paper were not unsettled in trusting their model more than the ^{14}C data by the fact that their radically new dating combined with noble gas temperatures yields very surprising paleoclimatic implications.

In yet another related paper on the Carrizo aquifer [5], Castro and Goblet discuss the problem of the non-uniqueness of the calibration of numerical models by hydraulic head data, and show that by using the He data they can select one of four scenarios for the distribution of hydraulic conductivities in the aquifer. I perfectly agree that He concentrations are very useful constraints for groundwater models, but I find it disturbing that equally important ^{14}C data apparently have been disregarded. I also agree with the view that ^{14}C dating of groundwater is problematic [4], but our knowledge of the He production and transport in the crust is also incomplete. It certainly is an important improvement to include the He data in the calibration of a numerical groundwater model, but if the resulting travel times differ so strongly from ^{14}C ages as a comparison of the age scales of Castro and Goblet [4] and Stute et al. [3] indicates, I remain doubtful of the uniqueness of the model calibration. Indeed, a simpler analytical model used by Castro et al. [2] apparently was in better agreement with both the ^4He and ^{14}C data.

3. Conclusions

Numerical models are a great thing, but data are the primary source of information that we have and should be used as much as possible. Sometimes more can be learned from interpreting data with simple concepts rather than complex models. All the information on mantle ^3He contributions given by Castro [1] could have been derived by simple mixing calculations

using the primary He data (and plots such as Fig. 2 in [1], which incorrectly are said not to allow for the estimation of contributions of specific components). The only new information obtained from the model is the very high residence time for the downstream part of the aquifer, and this is at odds with the ^{14}C data. I feel more confident in using the original data, which tell me that the He in the Carrizo aquifer is dominated by an external flux with an isotope ratio R_{ext} just slightly higher than the ratio of $0.055 R_a$ observed in the total He excess.

Finally, what does all this mean for the flux of mantle He? Independent of the exact value of R_{ext} , the mantle/crust mixing calculation essentially depends on the value of R_c assumed for the crustal endmember. Since the He excess is more or less strongly influenced by the external flux, the production ratio in its source region would need to be known. In the absence of other information, I propose to use the “typical” crustal value of $R_c = 2 \cdot 10^{-8}$ or $0.014 R_a$. With this value, $R_m = 9 R_a$, and Eq. (3), the mantle He (^4He) contribution to the external flux lies between 0.5% and 1.5%, depending on the value for R_{ext} (between $0.055 R_a$ and $0.14 R_a$). Of course, the mantle ^3He contribution is much larger, between 70% and 90%, but one definitely does not need a numerical model to learn that. Large scale mantle ^3He flux estimates do not depend on the details of He production in single aquifers, but rather on the mean $^3\text{He}/^4\text{He}$ production ratio in the crust.

References

- [1] M.C. Castro, Helium sources in passive margin aquifers—new evidence for a significant mantle ^3He source in aquifers with unexpectedly low in situ $^3\text{He}/^4\text{He}$ production, *Earth Planet. Sci. Lett.* 222 (2004) 897–913.
- [2] M.C. Castro, M. Stute, P. Schlosser, Comparison of ^4He ages and ^{14}C ages in simple aquifer systems: implications for groundwater flow and chronologies, *Appl. Geochem.* 15 (2000) 1137–1167.
- [3] M. Stute, P. Schlosser, J.F. Clark, W.S. Broecker, Paleotemperatures in the south-western United States derived from noble gases in ground water, *Science* 256 (1992) 1000–1003.
- [4] M.C. Castro, P. Goblet, Noble gas thermometry and hydrologic ages: evidence for late Holocene warming in Southwest Texas, *Geophys. Res. Lett.* 30 (24) (2003) 4.
- [5] M.C. Castro, P. Goblet, Calibration of regional groundwater flow models: working toward a better understanding of site-specific systems, *Water Resour. Res.* 39 (6) (2003) 25.