Report

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NOBLE GAS DATA FROM THE GREAT ARTESIAN BASIN PROVIDE A TEMPERATURE RECORD OF AUSTRALIA ON TIME SCALES OF 10^5 YEARS

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Abstract

We report noble gas data from 13 artesian wells in the south-western part of the Great Artesian Basin (GAB) in Australia. Atmospheric noble gases dissolved in groundwater allow the past soil temperatures of the recharge area to be reconstructed and radiogenic noble gas abundances yield estimates of the groundwater residence time. All measured samples have large excesses of He relative to atmospheric solubility equilibrium. Radiogenic ^4He concentrations, which were calibrated against ^{81}Kr groundwater ages, yield groundwater residence times up to 550 kyr. The noble gas temperatures calculated from the atmospheric noble gas concentrations show clear fluctuations of the soil temperature in Australia during the last 550 kyr. These results suggest that the noble gas temperature method can be applied to reconstruct continental paleoclimate conditions on time scales up to a few hundred kyr.

1. INTRODUCTION

Two key processes control the abundance of the noble gases, helium (He), neon (Ne), argon (Ar), krypton (Kr) and xenon (Xe) in groundwaters: *(1)* the *dissolution of atmospheric noble gases,* and (2) the *accumulation of radiogenic noble gas isotopes,* that are produced in relation to nuclear disintegration of radioactive nuclides. Dissolution of atmospheric noble gases reflects the climatic conditions at the water table prevailing during groundwater recharge, i.e. the soil temperature. On the other hand the amount of radiogenic noble gases points at least roughly to the time passed since a water parcel entered a groundwater system. Hence, in principle, the presence of atmospheric and radiogenic noble gases in groundwaters provides a unique temperature record to reconstruct paleoclimate conditions in continental areas.

In contrast to surface waters, atmospheric noble gases in groundwaters are generally found to be in excess relative to the expected equilibrium. The processes responsible for the formation of this excess component still remain vague, although several studies show that the excess is of purely atmospheric origin (excess air) and has a similar elemental composition as atmospheric air [1-3]. Observed deviations from atmospheric composition have been interpreted in terms of

elemental fractionation during non-equilibrium gas exchange at the groundwater table [4]. The concentrations of atmospheric noble gases are determined by the five parameters temperature, pressure, salinity, amount of excess air and possible fractionation. Since salinity and pressure (i.e. the altitude of the recharge area) at the water table are basically known, three free parameters remain. Recently new inverse models describing atmospheric gas dissolution in groundwater have been developed that determine the controlling parameters out of the measured noble gas concentrations using statistical optimisation routines [5, 6]. As soil temperature directly constrains atmospheric noble gas dissolution in groundwater [7], at present the noble gas method is the most quantitative approach to determine local paleotemperatures on continents. On large time scales well above the range accessable by ^{14}C groundwater dating, dispersive mixing may smooth gradients and might interfere with temperature determination using atmospheric noble gases. Noble gas temperatures (NGTs) derived from noble gases in groundwaters have up to now only been successfully used to reconstruct the temperature shift between the last ice age and the Holocene [4, 8- 10].

We report noble gas data and NGTs from 13 artesian wells in the south-western part of the Great Artesian Basin (GAB) in Australia in order to explore whether the noble gas method can be applied on time scales up to several 10^5 years. Samples were taken in 1998 during two expeditions within an IAEA research program focusing on the application of new tracer methods in old groundwater systems. During the first expedition 4 wells were sampled towards the south-eastern tip of the investigated area, whereas the second expedition visited 9 wells located further north, along the presumed north-western direction of groundwater flow. Samples from the first expedition (labeled with #1 to #4 in Figs. 1, 2 and 3) were taken from locations that are not necessarily interconnected hydraulically, especially sample #1 seems to originate from a different groundwater horizon of the GAB system. ^{81}Kr activity was analysed in all 4 samples of the first expedition to determine the local groundwater residence time [11].

2. RESULTS

2.1. Noble gases

All measured samples have large excesses of He relative to atmospheric solubility equilibrium. The He concentrations tend to increase along supposed groundwater flow lines. Concentrations reaching up to 10^{-4} cm^3STP/g (Fig. 1) are generally higher in the central part of the investigated area whereas lower concentrations are found closer towards the edge. Due to excess air Ne is supersaturated up to 100% relative to the expected atmospheric equilibrium. Interestingly, the Ne excess (as measure of the excess air component) decreases systematically with rising He concentrations. In some sample the noble gas abundance of the excess air component is not equal to the elemental composition of atmospheric air. However, the concentrations of atmospheric noble gases (Ne, ^{36}Ar, Kr and Xe) can be translated to noble gas temperatures (NGTs) in a statistically significant way, if re-equilibration between dissolved noble gases and trapped soil air is assumed [12]. Generally, the calculated NGTs reflecting the water temperature during recharge are higher than the regional recent annual mean air temperature of 22°C (Fig. 1). NGTs reach a first maximum of about 26°C at a ^4He concentration of $4 \cdot 10^{-5}$ cm^3STP/g. As He concentrations increase further to $8 \cdot 10^{-5}$ cm^3STP/g NGTs drop to a temperature minimum. At even higher ^4He concentrations NGTs rise again. If He concentrations are interpreted as qualitative estimates of groundwater age, NGTs seem to indicate at least two climatic cycles with a maximum temperature shift of about 4°C (Fig. 1).

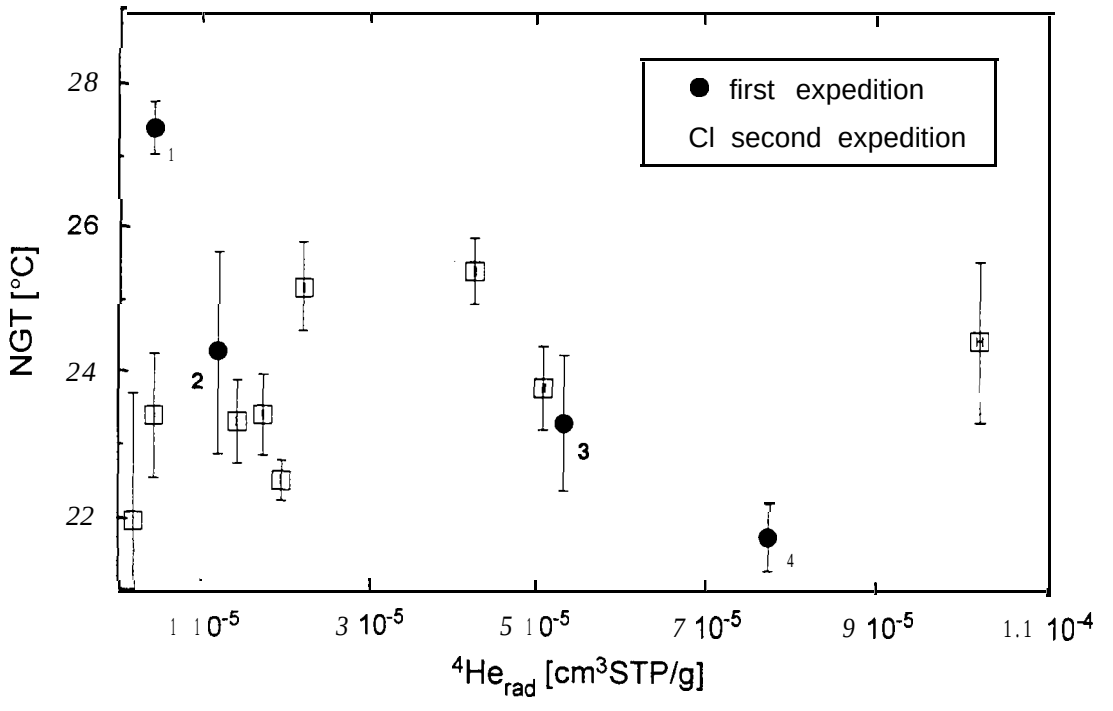

FIG. 1. Noble gas temperature (NGT) calculated from dissolved atmospheric noble gases plotted against the radiogenic ^4He concentration.

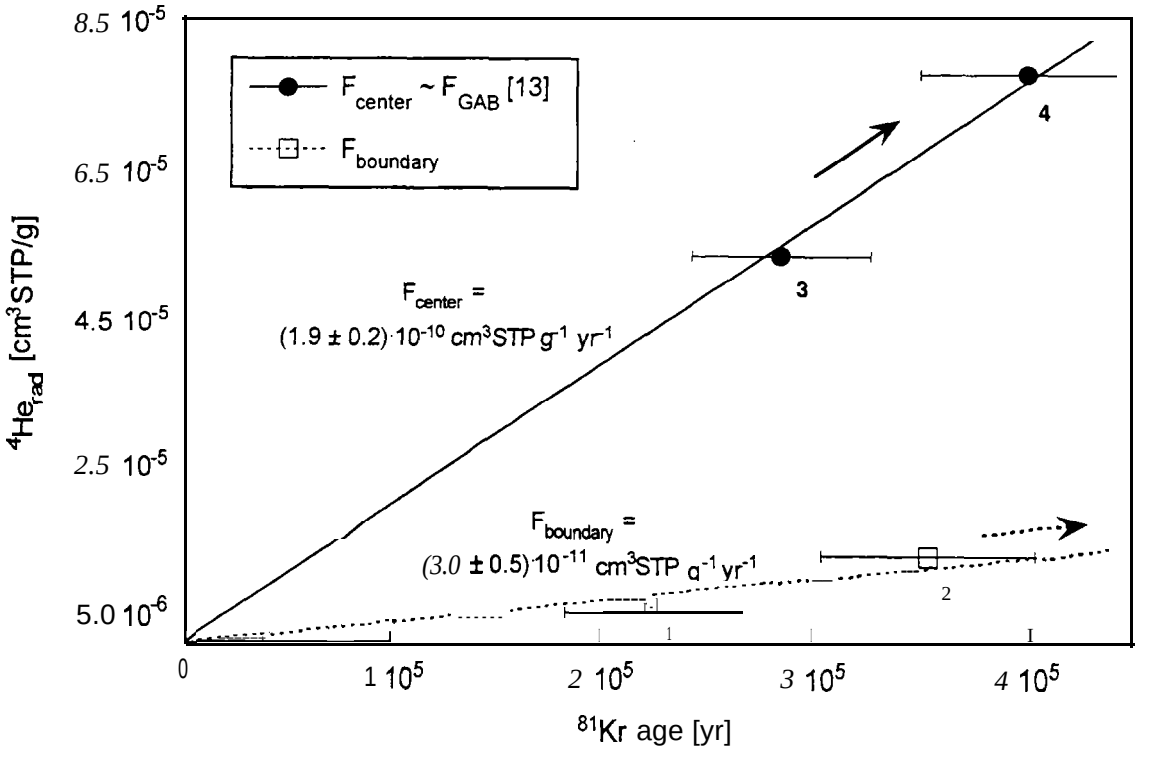

FIG. 2. Accumulation rates of radiogenic 'He.

3. DISCUSSION

3.1. Radiogenic ^{4}He and ^{81}Kr ages

Since ^{81}Kr groundwater ages are available for the four samples of the first expedition [11] they can be used to convert the measured ^{4}He concentrations into ^{4}He accumulation rates by dividing the ^{4}He concentration by the ^{81}Kr groundwater age (Fig. 2). All ^{81}Kr dated groundwater samples have water ages in the range between 200 and 400 kyr. Although the ^{81}Kr water ages vary only within a factor of two, the measured ^{4}He concentrations cover an order of magnitude, indicating that the ^{4}He accumulation rate is not homogeneous. The four samples form two clusters with different ^{4}He accumulation rates. Samples #3 and #4 from the center of the investigated area accumulate ^{4}He at a rate F_{center} of $(1.9 \pm 0.2)\cdot 10^{-10}$ $cm^3STP\cdot g^{-1}\cdot yr^{-1}$, whereas towards the western boundary (#1 and #2) the ^{4}He accumulation rate $F_{boundery}$ of $(3.0 \pm 0.5)\cdot 10^{-11}$ $cm^3STP\cdot g^{-1}\cdot yr^{-1}$ is about 6 times smaller (Fig. 2). The higher accumulation rate agrees within 30% with published estimates on the ^{4}He accumulation rate of the central region of the GAB [13]. The lower accumulation rate is one order of magnitude lower than previous estimates but still exceeds the in situ ^{4}He production of the GAB aquifers by almost a factor of 10 [13]. Whether our results reflect real differences of ^{4}He accumulation rates or whether the differences measure geological and/or geometrical heterogeneities of the GAB aquifer setting remains open and demands further investigations.

Since there are indications that at least one sample (#1) most probably does not originate from the same aquifer horizon as samples #3 and #4. Therefore and as the higher accumulation rate agrees with the characteristic GAB rate, we prefer to use the higher ^{4}He accumulation rate F_{center} to convert He concentrations into '^{4}He ages' (see Fig. 3). Although such a rough approach must be regarded as preliminary and hence the deduced '^{4}He ages' remain highly speculative guesses about the water residence time, the ^{4}He concentrations are the best age indicators for those water samples that have not been dated by means of ^{81}Kr. If the measured ^{4}He concentrations are interpreted as 'semi quantitative' ages the water samples from the investigated site have residence times up to 550 kyr (Fig. 3).

3.2. Noble Gas Temperatures (NGTs)

To our knowledge for the first time atmospheric noble gas concentrations were measured in GAB groundwaters that can be used to reconstruct significant average soil temperatures prevailing during groundwater recharge. Compared to former studies reporting in situ water temperatures up to the boiling point it is noticeable that the observed in situ groundwater temperatures of the investigated area are below 60°C. The successful NGT determination proves that the GAB aquifer system in principle provides a most valuable groundwater archive to reconstruct temperature evolution of the Australian continent, if the groundwater temperatures are low enough to prevent water samples from degassing during sampling.

It is evident that the aforementioned transformation of the measured ^{4}He concentrations into qualitative '^{4}He water ages' does not change the intrinsic temporal structure of the NGT record because the '^{4}He ages' are directly proportional to the determined ^{4}He accumulation rate, i.e. the individual ^{4}He abundances and ^{81}Kr ages of sample #3 and #4. Only samples #1 and #2 are shifted because their real groundwater ages as determined by their ^{81}Kr activity are much larger than the deduced '^{4}He ages' since these 'edge' samples seem to accumulate ^{4}He at a much smaller rate than 'common' GAB waters. The three samples (#2, #3 and #4) that have been dated by ^{81}Kr and which seem to originate from the same aquifer level (#1 is excluded from further discussion

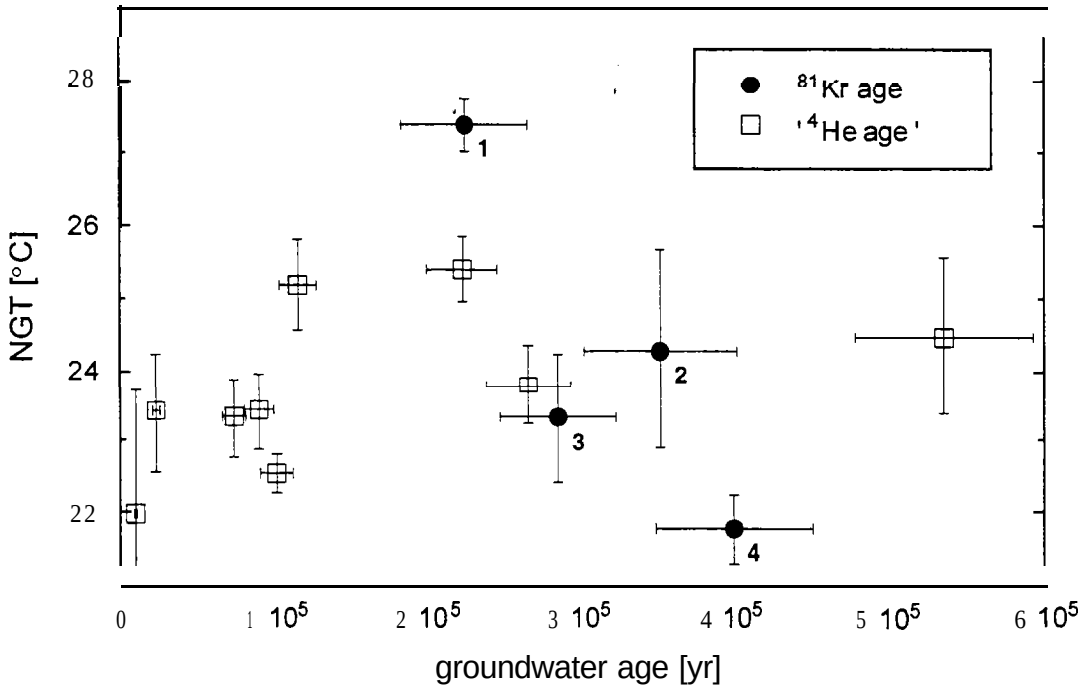

FIG. 3. Noble gas temperature (NGT) fluctuations of Australia as probed by concentrations of atmospheric noble gases during the last 600 kyr. The error of the calculated '^{4}He age' does not take into account large scale variations in the ^{4}He flux (see text for further explanations).

due to its different origin) mark a trend of NGT warming between 400 and 200 kyr before present (Fig. 3). The same warming phase was already present in the ^{4}He record at a concentration range between $4{\cdot}1\ 0^{-5}$ and $8{\cdot}1\ 0^{-5}$ cm^{3}STP/g (Fig. 1). The well dated samples follow basically the same tendency as the whole ensemble and hence pronounce the intrinsic structure of the temperature record. This fact adds some confidence to the attempt to interpret ^{4}He concentrations as measures for groundwater residence times.

However, since no recent groundwater samples were measured (i.e. samples containing no or only negligible radiogenic ^{4}He) the relations between calculated noble gas temperatures, soil temperatures and atmospheric temperatures remain unknown. It would be highly desirable to take groundwater samples near the recharge area in a future sampling expedition in order to calibrate the noble gas temperature record against the recent temperature conditions.

Temperature proxies with high temporal resolution, such as δ^{18}O records from marine foraminifera or from ice cores, report for the last 400 kyr 4 large climatic cycles during which the temperatures changed between warm interglacial and cold glacial conditions [14]. The time resolution of our GAB record is much too coarse that the presence of single glacial-interglacial cycles on the Australian continent could be proven. On the other hand the bottom line is that atmospheric noble gas data indicate significant temperature variations within central Australia during the last 600 kyr although at present the marginal time resolution prevents any assessment of the climatic dynamics and/or its rate of change.

4. CONCLUSIONS

We report noble gas concentrations from 13 artesian groundwater wells located in the south-western part of the Great Artesian Basin of Australia.

(1) The in situ groundwater temperatures are below 60°C and hence are much colder than temperatures reported in former studies. The low temperatures prevent (noble) gases from degassing during sampling. The received noble gas data finally allow interpretation of the atmospheric noble gas concentrations in terms of climatic conditions prevailing during recharge and interpretation of the radiogenic ^4He concentration in terms of qualitative groundwater ages.

(2) All samples have large ^4He excesses that generally tend to increase in direction of a supposed groundwater flow towards the central part of the investigated area. ^{81}Kr water ages [11] prove that radiogenic noble gases do not accumulate at equal rates inside the area. The central ^4He accumulation rate of $(1.9 \pm 0.2)\cdot 10^{-10}$ $cm^3STP\cdot g^{-1}\cdot yr^{-1}$ agrees within 30% with previous estimates of GAB He accumulation whereas towards the western boundary ^4He accumulates much slower ($(3.0 \pm 0.5)\cdot 10^{-11}$ $cm^3STP\cdot g^{-1}\cdot yr^{-1}$).

(3) If ^4He concentrations are interpreted as measures of the groundwater residence times the reported record spans a time range up to 550 kyr. Our results indicate that the time frame for which atmospheric noble gas concentrations reliably show soil temperatures can be expanded up to a few hundreds kyr in those cases where groundwater ages can reasonably be estimated. Although caution has to be exercised in drawing final conclusions the NGTs attest for Australia at least two climatic cycles over the last 550 kyr during which soil temperatures fluctuated by at least 4°C.

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