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Identification of He sources and estimation of He ages in groundwater of the North China Plain

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ABSTRACT

Dissolved helium concentrations and $^3He/^4He$ ratios were measured for 18 groundwater samples collected from the Quaternary confined aquifers in the North China Plain (NCP). The dissolved helium concentrations ranged from 1×10^{-7} to 1×10^{-6} $cm^3STP \cdot g^{-1}$ in the 14 samples from the central plain, but was approximately two orders of magnitude higher, between 6×10^{-6} and 9×10^{-5} $cm^3STP \cdot g^{-1}$, in 4 samples from the coastal plain. Based on these concentrations and the corresponding $^3He/^4He$ ratios varying from 0.09 to 0.55 R_a (where R_a is the $^3He/^4He$ ratio of air), the dissolved helium in groundwater in the central plain was identified to be primarily a mixture of atmospheric helium with radiogenic helium and a representative radiogenic helium ratio was estimated to be 0.035 R_a. Despite the high fraction of terrigenic ^4He in the samples from the coastal plain, their $^3He/^4He$ ratios were found to be significantly above this radiogenic value, ranging between 0.20 and 0.37 R_a, indicating the presence of a mantle-derived He component in this area. About 2–4% mantle helium was estimated to be present in the groundwater of the coastal plain, which probably is associated with the regional Cangdong fault and tectonic activities. Based on the radiogenic He component, ^4He ages of the groundwater in the central plain were calculated by assuming either pure in situ production or an external helium flux J_0 of 4.7×10^{-8} $cm^3STPcm^{-2}a^{-1}$. The estimated ^4He ages fall between 9.5 and 51.4 ka and are comparable to the ^{14}C ages, suggesting that the results of ^4He dating are reasonable and can be an effective tool to estimate groundwater residence times under suitable conditions.

1. Introduction

Helium-4 is a stable isotope and a decay product of naturally occurring actinides, primarily uranium and thorium that are present in variable concentrations in all rock types (Mamyrin and Tolstikhin, 1984; Ozima and Podosek, 2002). Helium-4 enters groundwater from different sources including atmosphere, in situ production, inflow from the mantle and release from the aquifer sediments (Kazemi et al., 2006). Groundwater dissolves atmospheric He as a consequence of air-equilibration during recharge and thereafter accumulates radiogenic He which is generated within the aquifer and from adjacent layers of aquitards and/or an external He flux (Andrews and Lee, 1979; Lehmann et al., 2003; Tolstikhin et al., 2011). This process induces changes in the $^3He/^4He$ ratio and the concentration of dissolved He, which may be used to estimate the mixing proportions of atmospheric and radiogenic He in the groundwater.

The radiogenic He that gradually supplements the atmosphere-derived He can be applied to estimate groundwater residence time in confined aquifers (e.g., Savchenko, 1935; Andrews, 1985; Aeschbach-Hertig et al., 2002; Plummer et al., 2012). It has been shown that radiogenic ^4He is able to provide groundwater residence time estimates that are comparable with ^{14}C ages in simple aquifer systems (Castro et al., 2000). An advantage of helium dating compared with the traditional method of ^{14}C dating is that it can be used for estimating very old ages because He is stable and thus its applicability is not limited in time. ^4He can be used to estimate groundwater mean residence times ranging from 100 up to 1×10^8 a (Rübel et al., 2002; Tolstikhin et al., 2011), whereas ^{14}C dating is limited to about 5–6 half-lives corresponding to 30–35 ka. However, considerable scientific questions still remain concerning the applicability of ^4He, such as the separation of the individual He components and determination the external He flux from the crust and mantle.

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Although the radiogenic or crustal He is often the only non-atmospheric He component in groundwater, in some cases significant contributions of He from the Earth's mantle have been found in aquifers (e.g., Stute et al., 1992; Kulongoski et al., 2005). These two terrigenic components can be distinguished based on their $^3He/^4He$ ratios, about three orders of magnitude higher in the mantle than in the crust (Mamyrin and Tolstikhin, 1984). The presence of mantle He in aquifers in shallow layers of the crust implies that routes for the migration of mantle fluids to the surface exist and is often associated with tectonic or volcanic activity. Local high $^3He/^4He$ anomalies may be related to fracture zones with locally enhanced permeability and have been used to identify zones with high potential for geothermal energy development (Kennedy and van Soest, 2007).

The purposes of this paper are to identify the sources of helium components in groundwater of the North China Plain (NCP) as well as to evaluate their contributions to the total He inventory, and thus to estimate groundwater age in the deep confined aquifers of the NCP, which is significant for the assessing groundwater resources renewability and therefore making measures for water resources management under the extensive exploitation in NCP.

2. Hydrogeological setting

The NCP is situated in the eastern part of China, it extends from the Taihang mountains in the west to the Bohai sea in the east and from the Yanshan Fold in the north toward the Yellow River in the south (Chen et al., 2003; von Rohden et al., 2010). It is one of the major food producing areas of China and is densely populated with more than 100 million inhabitants. The majority of water resources consumed for domestic, industrial and agricultural purposes are supported by groundwater, and this has resulted in a rapid decline of water levels in recent decades and the development of several big depression cones (Zhang et al., 2000).

The NCP is a thick Cenozoic sedimentary basin and it can be separated into three subsystems as the piedmont fluvial plain, the central alluvial and flood plain, and the coastal delta plain on the basis of topography and aquifer system characteristics (Fig. 1). There is great difference spatially among the hydraulic characteristics of the Quaternary aquifer in the NCP. The Quaternary sediments are 150–600 m thick and consist of unconsolidated deposits with clay, silty clay, sand and sandy gravel layers, where the clay and silty clay constitute relatively impermeable layers as aquitards.

The NCP hydrogeological framework has been extensively studied and consists of 4 water-bearing formations in vertical direction based on lithological and hydrodynamic properties, corresponding to the Holocene formation (Q_4), the upper Pleistocene (Q_3) formation, the middle Pleistocene formation (Q_2) and the lower Pleistocene formation (Q_1), respectively (Chen et al., 2003; Zhang et al., 2009).

This study focuses on the two important confined aquifers Q_2 and Q_1 located in the central and coastal plain of the NCP, which two aquifers are in hydraulic continuity and were strictly controlled for the groundwater exploitation. Groundwater flow in the confined aquifers is generally from the west to east (Zhang et al., 2009). Most recharge is from infiltration of precipitation near the outcrop area. Detailed descriptions of the aquifer system in the NCP have been provided (c.f. Zhang et al., 2000, 2009; Chen et al., 2003; Kreuzer et al., 2009).

3. Sampling and measurement

A total of 52 groundwater samples were collected along the piedmont to the east coastal plain during two campaigns that took place in 2004 and 2005. This project was carried out by Chinese researchers (IHEG) in cooperation with German researchers (IUP). Some data have been reported before in a paleoclimate study (Kreuzer et al., 2009) and a study on the recharge regime (von Rohden et al., 2010). This paper focuses on the 18 noble gas samples which were collected from the deep confined aquifers along the central to the coastal plain, and aims to identify the sources of helium dissolved in groundwater as well as to estimate mean residence time with radiogenic ^4He.

All samples were collected after three to five well-bore volumes had been purged, and after Eh, pH, and temperature readings had reached constant values. The samples could be characterized into 5 groups listed in Table 1 on the basis of hydrogeological setting and their respective $^3He/^4He$ ratios. In particular, water samples for noble gases analysis were directly transferred and sealed off in copper tubes, and analyzed at the Institute of Isotope Geochemistry and Mineral Resources, ETH Zurich, using mass spectrometry with typical precisions of ± 0.7% for He, ± 1.1% for Ne, ± 0.8% for Ar, ± 1.2% for Kr, ± 1.6% for Xe, and ± 0.7% for the $^3He/^4He$ ratio.

4. Methods

4.1. Sources of helium in the groundwater

The concentrations of He isotopes (^3He, ^4He) in groundwater usually are in excess of those expected for water in solubility equilibrium with the atmosphere (Heaton and Vogel, 1981; Stute et al., 1992; Wilson and McNeill, 1997; Castro et al., 2000). These excesses can result from 5 different sources: (1) an excess air component (Heaton and Vogel, 1981); (2) tritiogenic ^3He, the β-decay product of natural and bomb-produced ^3H, which can provide age information on relatively young groundwater systems (Schlosser et al., 1988; Visser et al., 2014) ;(3) nucleogenic ^3He, originating from the $^6Li(n, \alpha)^3H(^3He)$ reaction; (4) radiogenic ^4He, the result of α-decay of the natural U and Th decay series isotopes in common rocks; and (5) mantle contributions to both ^3He and ^4He. Therefore, the measured helium concentrations in groundwater samples can be interpreted as the sum of these components (Torgersen, 1980; Weise and Moser, 1987). Each component has a characteristic $^3He/^4He$ ratio, for example, the Earth's atmospheric air ratio R_a of 1.384×10^{-6} (Clarke et al., 1976), the typical crustal (radiogenic and nucleogenic) helium isotope ratio of 2.0×10^{-8} or 0.02 Ra (Mamyrin and Tolstikhin, 1984), and the typical mantle-derived helium isotope ratio of 1.2×10^{-5} or 8 Ra (Craig and Lupton, 1981; Ozima and Podosek, 2002; Kazemi et al., 2006).

4.2. Modeling of excess air

In most natural groundwater, the concentration of dissolved noble gases is significantly higher than expected from equilibrium with atmospheric air. This phenomenon has been termed “excess air” and interpreted as the result of dissolution of small air bubbles trapped in groundwater by fluctuations of the groundwater table (Heaton and Vogel, 1981; Kipfer et al., 2002). Several models such as the total dissolution or unfractionated air (TD or UA) model, the partial diffusive re-equilibration (PR) model, and the closed-system re-equilibration (CE) model have been developed to explain and quantitatively describe this excess air component (Aeschbach-Hertig et al., 2000; Aeschbach-Hertig and Solomon, 2013). Inverse modeling algorithms have been developed to estimate the model parameters and study the ability of the different models to fit observed data (Aeschbach-Hertig et al., 1999). Among the mentioned models, the CE model was recognized to be physically realistic and to provide the best fit for interpreting the noble gas data in many case studies (Aeschbach-Hertig et al., 2000; Klump et al., 2007). The basic assumption of the CE model is that