



Using helium and other noble gases in ocean sediments to characterize active methane seepage off the coast of New Zealand



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ABSTRACT

Recently developed analytical techniques to determine the abundances of noble gases in sediment pore water allow noble-gas concentrations and isotope ratios to be measured easily and routinely in lacustrine sediments. We applied these techniques for the first time to ocean sediments to investigate an active cold methane seepage system located in the South Pacific off the coast of the North Island of New Zealand using ³He/⁴He ratios determined in the sediment pore water.

The results show that more ³He-rich fluids are released in the vicinity of the Pacific–Australian subduction zone than at the forearc stations located closer to the New Zealand coast. However, the He isotope signature in the sediment column indicates that only a minor part of the He emanating from deeper strata originates from a depleted mantle source. Hence, most He in the pore water is produced locally by the radioactive decay of U and Th in the sediment minerals or in the underlying crustal rocks. Such an occurrence of isotopically heavy crustal He also suggests that the source of the largest fraction of methane is a near-surface geochemical reservoir. This finding is in line with a previous $\delta^{13}\text{C}$ study in the water column which concluded that the emanating methane is most likely of biological origin and is formed in the upper few meters of the sediment column. Moreover, the prevalence of isotopically heavy He agrees well with the outcome of other previous studies on island arc systems which indicate that the forearc regions are characterized by crustal He emission, whereas the volcanic arc region is characterized by the presence of mantle He associated with rising magma.

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1. Introduction

Noble gases make ideal tracers for studying the transport of other dissolved species in the pore water of unconsolidated sediments (Brennwald et al., 2005; Strassmann et al., 2005; Tomonaga et al., 2011a; Brennwald et al., 2013).

Henry's Law describes the equilibrium partitioning of noble gases between air and water. As the gas-specific Henry coefficients are controlled by the temperature and salinity of the water, the solubility equilibrium concentrations of dissolved noble gases directly reflect the physical conditions that prevailed when the water was last in contact with the atmosphere. This straightforward physical concept underlies the use of the concentrations of dissolved noble gases to determine mixing and deep-water exchange in lakes and oceans, to reconstruct past climate conditions from groundwater, and to analyze past hydraulic conditions (for recent reviews see Kipfer et al., 2002; Schlosser and

Winckler, 2002; Aeschbach-Hertig and Solomon, 2013; Stanley and Jenkins, 2013).

In surface waters, advection and turbulent mixing affect the transport of heat and solutes in the same manner (e.g., Schwarzenbach et al., 2003). Hence, concentrations of dissolved noble gases in lakes and in the oceans have been found to agree closely with the expected atmospheric equilibrium concentrations determined from the ambient atmospheric pressure and the local temperature and salinity of the surrounding water mass (e.g., Aeschbach-Hertig et al., 1999; Peeters et al., 2000; Kipfer et al., 2002).

During sedimentation, water from the sediment/water interface is incorporated into the growing sediment column. Atmospheric noble-gas concentrations in the pore water of sediments are, therefore, expected to agree with the noble-gas concentrations of the overlying water mass at the time when the sediment was deposited (Strassmann et al., 2005). Consequently, deviations of the noble-gas concentrations measured in the pore water from the expected atmospheric equilibrium concentrations can be used to identify key physical processes controlling the accumulation or loss of dissolved species in the pore space (for a review see Brennwald et al., 2013).

In particular, as helium (He) emanates from the solid earth into the atmosphere (O'Nions and Oxburgh, 1983; Mamyrin and Tolstikhin,

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1984; Oxburgh and O'Nions, 1987), the origin of other solutes emanating from the same geogenic reservoir can be determined in a straightforward manner using the He concentrations and isotope ratios measured in the sediments (Barnes, 1987; Holzner et al., 2008; Lan et al., 2010; Tomonaga, 2010; Tomonaga et al., 2011a). Three major geochemical He reservoirs are commonly distinguished by their characteristic He isotope signature: the atmosphere ($^3\text{He}/^4\text{He} \approx 1.384 \cdot 10^{-6}$, Clarke et al., 1976), the Earth's mantle ($^3\text{He}/^4\text{He} \approx 10^{-5}$), and the Earth's crust ($^3\text{He}/^4\text{He} \approx 10^{-8}$). Hence, the He isotope ratios in aquatic systems allow straightforward assessment of the occurrence of fluids originating from the deep lithosphere (Kipfer et al., 2002; Schlosser and Winckler, 2002).

The development of a method for the sampling and determination of noble gases in sediment pore water by Brennwald et al. (2003) allowed noble-gas concentrations in unconsolidated lake sediments to be measured in a robust and experimentally feasible manner for the first time. A major experimental problem encountered when applying this method to ocean sediments is that complete extrusion of the sediment samples from the sampling container cannot always be achieved. Sediments of very fine texture and relatively low porosity ($\leq 60\%$) are especially prone to partial extrusion, with part of the bulk sediment remaining in the form of compact clusters in the copper tube used as a sample container. As the presence of these clusters severely hampers and attenuates the degassing process, noble-gas extraction is often incomplete. Moreover, the method of Brennwald et al. (2003) is known to liberate He from the sediment grains in response to the intense heating of the bulk sediment samples that is part of the extraction process. Until now, it has therefore been necessary to compare the He abundances measured in the pore water with the He concentrations and isotope ratios in the overlying water column to distinguish the He released locally from the sediment matrix from the terrigenous He migrating through the sediment column.

A recent improvement to the analytical method of determining noble-gas abundances in the pore water of unconsolidated sediments (Tomonaga et al., 2011b) shows that the known experimental limitations of the method described by Brennwald et al. (2003) can be overcome if the pore water is completely separated from the sediment matrix by centrifuging the bulk sediment samples within the ultra-high-vacuum sealed copper tubes.

In this study we adapted the centrifuging method of Tomonaga et al. (2011b) to determine noble-gas concentrations in the pore water of ocean sediments acquired in the vicinity of an active cold methane (CH_4) seepage system off the coast of the North Island of New Zealand (Faure et al., 2006, 2010; Greinert et al., 2010; Linke et al., 2010; Naudts et al., 2010; Sommer et al., 2010). To our knowledge this is the first published study in which a comprehensive analysis of all noble-gas species (i.e., He, Ne, Ar, Kr, and Xe), including the isotope ratios $^3\text{He}/^4\text{He}$, $^{20}\text{Ne}/^{22}\text{Ne}$, and $^{36}\text{Ar}/^{40}\text{Ar}$, has been carried out in the pore water of open-ocean sediments.

In the Black Sea, noble gases have been found to yield valuable information about the origin of methane injected into the water column by high-intensity seeps. The determination of noble-gas abundances allowed the functional and tectonic relationship between CH_4 emission and the activity of mud volcanoes to be studied (Holzner et al., 2008). In the work by Holzner et al. (2008) noble gas concentration profiles in the water column and in the sediment pore water at active seep sites revealed prominent anomalies in relation to CH_4 emission. Super-saturations of the light noble gases (He and Ne) observed relatively close to the sea floor were interpreted as the result of gas exchange between the water and the rising bubbles. Isotopically heavy He in the pore water close to an active mud volcano identified the crustal origin of the locally emitted fluids, whereas in other regions of the Black Sea also traces of mantle-type He were found.

Based on the insights gained on the methane emission in the Black Sea using noble gases, in this study we aim to identify the geochemical origin of methane at cold seeps in the South Pacific, in particular by

using He isotopes to determine the origin of the ascending fluids (see Faure et al., 2006). Our work completes and extends the research conducted by other groups during the same cruise on *RV SONNE* (Faure et al., 2010; Greinert et al., 2010; Linke et al., 2010; Naudts et al., 2010; Sommer et al., 2010) to study the fluid emission at the Hikurangi Margin off shore New Zealand.

2. Regional setting

New Zealand lies between the Australian and Pacific tectonic plates. To the north-east of New Zealand the Pacific Plate is being subducted below the Australian Plate at the Hikurangi Margin leading to the development of an island-arc system (Cole and Lewis, 1981). To the south of New Zealand the Australian Plate is being subducted under the Pacific Plate at the Puysegur Margin. Plate movement results in volcanic and large seismic activity in the North Island of New Zealand. Within the South Island the Alpine Fault, a well developed transform fault, marks the plate boundary (Fig. 1).

For noble-gas analysis, three sediment cores were taken in March 2007 during the *RV SONNE* Cruise 191 lead by IFM-GEOMAR in the vicinity of the Pacific–Australian subduction zone in the south-west Pacific, off the coast of the North Island of New Zealand (Fig. 1). The region, stretching along the gas-hydrate-bearing Hikurangi Margin, is characterized by the emanation of cold, methane-rich fluids released from the sediments into the water column (Faure et al., 2006, 2010; Greinert et al., 2010; Linke et al., 2010; Naudts et al., 2010). This cold seepage environment also functions as the habitat for specific biological communities that interact with the seep geochemistry (Sommer et al., 2010).

Previous investigations of the He isotope composition of gas samples acquired in the continental region of New Zealand in relation to the emission of CO_2 and CH_4 have extensively documented the spatial distribution of He emission in this region (Sano et al., 1987; Giggenbach et al., 1993). According to these studies, while mantle He seems to be typical for the volcanic arc and back-arc regions ($^3\text{He}/^4\text{He} \approx 10^{-5}$), the forearc region is characterized by the presence of radiogenic He produced in the crust ($^3\text{He}/^4\text{He} \leq 10^{-7}$). A similar geographical distribution of the He isotope signature observed in northeastern Japan (Sano and Wakita, 1985; Sano and Nakajima, 2008) seems to be characteristic of island arc systems (e.g., Horiguchi et al., 2010).

The first core for noble-gas analysis was taken at the “Rock Garden” station ($40^\circ 1.939' \text{ S}$, $178^\circ 9.654' \text{ E}$) in the immediate vicinity of the subduction zone at a water depth of 660 m. As the name of this station implies, the presence of rocks allowed only small cores with a maximum length of 1 m to be taken. Inspection of the “Rock Garden” site using a remotely operated vehicle (ROV) showed active macroscopic bubble emission at the sediment/water interface (Naudts et al., 2010).

The other two sampling stations, “Reference” ($40^\circ 1.414' \text{ S}$, $177^\circ 48.953' \text{ E}$; water depth of 1180 m) and “Bear's Paw” ($40^\circ 3.195' \text{ S}$, $177^\circ 49.188' \text{ E}$; water depth of 1100 m), are located at Omakere Ridge, closer to the shore of New Zealand (for additional maps see Greinert et al., 2010; Linke et al., 2010). The distance between the stations at Omakere Ridge and “Rock Garden” is about 30 km, “Reference” and “Bear's Paw” are 3 km apart from each other.

3. Methods

The samples for noble-gas analysis were acquired on *RV SONNE* using our standard sampling technique (Brennwald et al., 2003). Immediately after core recovery on the ship, the bulk sediment was transferred into small copper tubes (inner diameter $\approx 0.6 \text{ cm}$, length $\approx 50 \text{ cm}$) by squeezing the sediment core between two pistons moving along the axis of the plastic liner (see Brennwald et al., 2003). The copper tubes, which were attached to the side of the liner by Swagelok fittings, were filled with bulk sediment and sealed airtight by closing two metal clamps to avoid air contamination and degassing (Beyerle et al., 2000; Brennwald et al., 2003).

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