Mantle helium in pore fluids of deep-sea sediments around the Azores archipelago

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ABSTRACT

We present noble gas concentrations and helium isotope ratios measured in the pore fluids of two deep-sea sediment cores taken at the Azores Plateau. Ne—Xe concentrations were used to estimate the atmospheric noble gas component within the pore fluids and to then determine the terrigenic helium component. The concentrations of terrigenic ^{3}He indicate the presence of hydrothermal fluids and a mantle He component within the sediment pore space. At both sampling locations, the concentration of terrigenic helium increases with sediment depth. The observed concentration gradients suggest a diffusive transport of He through the sediments. The helium isotope ratio of the source of the terrigenic helium is characterized by a typical mid-ocean ridge basalt (MORB). These results support previous studies suggesting that the pelagic sediments surrounding the Azores host active hydrothermal systems.

Our data indicate that the transport of terrigenic helium takes place diffusively over larger areas of the ocean floor, and that the concentration of terrigenic helium in the pelagic sediments increases towards the Mid-Atlantic Ridge and Terceira Rift.

The data further suggests that in the Azores region the terrigenic noble gas component is only detectable in the sediment pore fluids and cannot be observed in the overlying bottom water. This highlights the potential of measuring noble gas concentrations and isotope ratios in the sediment column to identify hydrothermal activity in the ocean.

1. Introduction

The Azores are a volcanically active archipelago located at the Mid-Atlantic Ridge (MAR) on a triple-junction of three diverging tectonic plates (North American, Eurasian and African plate). The Mid-Atlantic Ridge separates the islands in eastern and western part. In the north of the Azores, the Terceira Rift is extending south-east from the MAR (see Fig. 1), while in the south the East Azores Fractures Zone is extending East from the MAR. Tectonic and magmatic activity are closely linked in the Azores archipelago, demonstrated by the alignment of island volcanoes and seamounts along prominent tectonic lineaments and fault zones (Weiß et al., 2015a).

Despite frequent volcanic and seismic activity in this region, hydrothermal systems had initially only been found on land and in shallow water (Ferreira et al., 2005; Viveiros et al., 2010; Couto et al., 2015), and until recently there had been no evidence for hydrothermal activity in the deep ocean sediments around the archipelago. On land, degassing areas associated with active hydrothermal systems and degassing from soils are found mainly on the islands of S. Miguel, Terceira and Graciosa (Ferreira et al., 2005). Shallow-water hydrothermal vents have been reported on the flanks of the Azores islands (Couto et al., 2015). The best known example for this shallow-water hydrothermal activity is the D. Joao de Castro seamount with several vents, located between the islands of Terceira and S. Miguel only 20 m below sea level (Cardigos et al.,

2005). However, since the discovery of the seamount, the hydrothermal system seems to have ceased in activity and more recent expeditions found no signs of hydrothermal activity (Beier, 2016).

Some first evidence that the deep seafloor around the Azores might be a hydrothermally active region was the discovery of volcanic cones connected to faults in the eastern Terceira Rift, which could potentially host hydrothermal systems (Weiß et al., 2015b). Close to these cones, Schmidt et al. (2019) found Mg, SO₄, and alkalinity anomalies in pore fluids of near-surface deep-ocean sediments, which were speculated to be evidence for waning hydrothermal activity. Additionally, major element composition data from pore waters of deep-sea sediments from different locations around the Azores Plateau indicates the existence of submarine hydrothermal systems located far off the MAR (Schmidt et al., 2020).

However, despite these first indications for hydrothermal activity in the pelagic sediments of the Azores region, little is known about the transport mechanisms of these (potentially) hydrothermal fluids through the sediments.

The knowledge about fluid evolution and their dynamics in ocean sediments hosting hydrothermal systems is in general very limited. Up to now only few sediments bearing hydrothermal fluids are described worldwide, e.g., in the Guyamas Basin or at Loki's Castle (Diehl and Bach, 2020). Compared to black smoker systems, hydrothermal fluids emanating through sediments show very variable chemical compositions due to the interaction with the surrounding sediments (e.g. Baumberger et al., 2016; Devey et al., 2021). The questions on how hydrothermal fluids migrate through ocean sediments and how the fluid release through sediments affects the element cycles in the ocean are - as a result of the experimental difficulties to adequately determine fluid dynamics in sediments - hardly addressed and call for investigation.

In this study, we use noble gas concentrations and helium isotope ratios from pore water of pelagic sediments around the Azores to identify the presence of mantle fluids. The aim of this work is to provide supporting evidence that the ocean floor around the Azores is indeed hydrothermally active and to shed light on the question how fluid migration takes place in such a sediment-hosted hydrothermal system.

2. Methods

Noble gases are excellent tracers to identify atmospheric fluid components, as the concentrations of Ne, Ar, Kr and Xe in surface water are only depending on the physical conditions prevailing during air/water equilibration with the atmosphere, such as water temperature, salinity and air pressure (Kipfer et al., 2002; Brennwald et al., 2013), and as their atmospheric mixing ratios are known to be constant over geological timescales. Due to the synchronous transport (e.g. advection/turbulent diffusion) of noble gases, salt and heat in the ocean, bottom water concentrations of noble gases depend on local bottom water temperature and salinity (Brennwald et al., 2013). During sedimentation, the overlying bottom water is incorporated into the sediment column, so the noble gas concentrations in sediment pore water are often equal to the concentrations in air-saturated (ocean) water (Brennwald et al., 2003; Strassmann et al., 2005; Tomonaga et al., 2011a). Thus, noble gases can be used to reconstruct the environmental conditions during pore water entrapment (e.g. Tomonaga et al., 2014; Brennwald et al., 2004) and to determine atmospheric and non-atmospheric components, e.g. in the case of this study to determine whether a terrigenic He component is present in the pore space. Noble gas analysis in the pore fluids of ocean sediments has already been employed successfully to identify a mantle helium component in the sediments, e.g. close to the Pacific–Australian subduction zone off the coast of New Zealand (Tomonaga et al., 2013), along the Nankai Trough close to Japan (Tomonaga et al., 2020) or in the vicinity of an active hydrothermal system in the Guaymas Basin, Gulf of California (Horstmann et al., 2021).

In sediments terrigenic ^{4}He accumulates from the radioactive decay of U and Th in the pore space over time and can be used to estimate pore water ages (Mamyrin and Tolstikhin, 1984; Solomon, 2000; Kipfer et al., 2002). Fluids enriched in ^{3}He with respect to the atmospheric helium isotope ratio hint to a helium source derived from the mantle, as the Earth's mantle is known to be enriched in isotopically light helium (Mamyrin and Tolstikhin, 1984; Porcelli and Ballentine, 2002). For the basaltic sediments forming new ocean floor at mid-ocean ridges, (mid-ocean ridge basalts, "MORB") the typical ^{3}He/^{4}He ratio is approximately 6.5–9.5 R_A (Graham, 2002), where 1 R_A represents the atmospheric ^{3}He/^{4}He ratio of $1.384 \cdot 10^{-6}$ (Clarke et al., 1976). On the Azores islands, helium isotope ratios observed in basalts or olivine crystals are usually found to agree with typical MORB values (Métrich et al., 2014; Moreira et al., 1999). However, on the island of Sao Miguel lower-than-MORB ^{3}He/^{4}He ratios have been reported (Moreira et al., 2012). Higher than MORB ^{3}He/^{4}He ratios have been observed in some samples from Pico and Sao Jorge (e.g. Moreira et al., 2018).

Samples for noble gas analysis were acquired during the *GEOMAR* cruise *M141/1* with the research vessel *Meteor* in the area of the Azores archipelago. We present noble gas concentrations in pore fluids of deep-
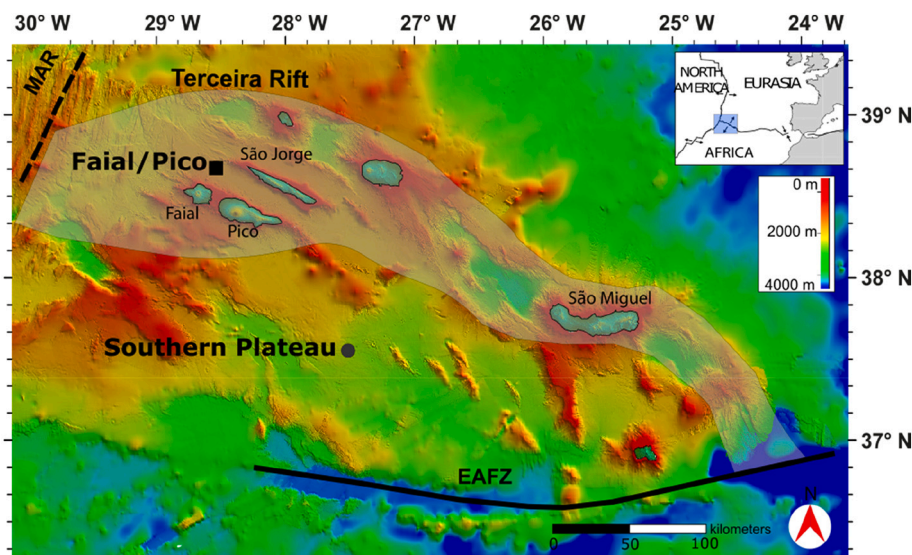

Fig. 1. Bathymetric map of the sampling region in the Azores Plateau, adapted from Schmidt et al. (2020). The grey area depicts the diffuse plate boundary of the Terceira Rift based on Marques et al. (2013); MAR: Mid-Atlantic Ridge, EAFZ: East Azores Fracture Zone.

sea sediments from two sites (see Fig. 1). The first sampling site (Southern Plateau) is located south of the Eastern Azores islands in a distance of about 80 km from the Terceira Rift and 200 km from the MAR. The second sampling site (Pico/Faial), located between the islands of Pico, Faial and Sao Jorge, lies on the Terceira Rift and in closer distance to the MAR (about 100 km). Ocean depths at the Southern Plateau and Pico/Faial sites were 2175 m and 1225 m, respectively. At both locations gradients in major element composition, i.e. increasing Ca, Sr and NH_4 concentrations and decreasing Mg, SO_4 concentrations and total alkalinity, have been observed in the sediment column, which is a pattern typically found for hydrothermally-derived fluids, however, the gradients are much stronger at Pico/Faial (Schmidt et al., 2020).

The sediment samples for noble gas analysis were taken and processed according to the well-established sampling protocols of Brennwald et al. (2003) and Tomonaga et al. (2011b). The two sediment cores were acquired by a gravity corer with liners of 5 m length. Prior to coring, holes were drilled along the liners and covered with tape during coring. After retrieving the cores, the liners were cut into 1 m sections, and copper tubes were attached to the previously drilled holes via stainless-steel fittings. A custom-made sediment press with two pistons was used to transfer the bulk sediment from the liner segments into the copper tubes, which were then sealed with metal clamps (for a detailed description and additional technical details of this sampling method, see Brennwald et al., 2003). Sediment samples for noble gas analysis were taken at intervals of 40–60 cm along the liners. The copper tubes containing the bulk sediment were processed for dissolved gas analysis in the pore water at *Eawag* in Switzerland according to the protocols established by Tomonaga et al. (2011b, 2014). The copper tubes containing the sediment samples were centrifuged to isolate the pore water from the sediment matrix. After centrifugation, a metal clamp was placed along the copper tube at the sediment/water interface to separate the free water and the sediment. For the subsequent noble gas analysis only the sample part containing the water phase was used.

At the Noble Gas Laboratory at ETH Zürich, noble gas analysis was performed with static mass spectrometry according to a well-established experimental protocols to quantitatively determine the concentrations and isotopic ratios of noble gases in water samples (for a detailed description of the experimental protocol, including gas separation and analysis, see Beyerle et al., 2000). An atmospheric air standard was used for calibration of the noble gas measurements. The concentrations of He, Ne, Ar, Kr, and Xe, as well as the ^{3}He/^{4}He isotope ratio in the sediment pore water were analyzed with an overall experimental error of <1.5% for He, Ne, Ar, and Kr, < 2.5% for Xe and < 5% for the ^{3}He/^{4}He isotope ratio. Equilibrium concentrations of noble gases in air-saturated ocean water were calculated using the solubility data set recommended by Kipfer et al. (2002). Additionally to noble gas measurements, U and Th concentrations in the dry sediment were determined for the top and bottom sample of each core at *Eawag* using γ-spectroscopy.

To obtain the terrigenic helium concentration in the samples, we determined the atmospheric component for all noble gases (see Kipfer et al., 2002) with the unfractionated air model:

$$C^i_{meas} = C^i_{ASW}(T, S, P) + C^i_{AE} + C^i_{terr}$$
$$= C^i_{ASW}(T, S, P) + A \cdot z^i + C^i_{terr} \qquad (1)$$

where C^i_{meas} is the measured concentration and C^i_{ASW} is the air-saturated water (ASW) concentration of gas i (i = He, Ne, Ar, Kr, Xe). C^i_{ASW} depends on water temperature T, salinity S, and atmospheric pressure P. The term C^i_{AE} denotes the concentration of gas i due to an atmospheric air excess that was observed in some samples. We note that the elemental composition of ASW and atmospheric air differs from each other and that pore water in ocean sediments is typically known to be in atmospheric equilibrium. Therefore the observed supersaturation of noble gases in some of the samples can be interpreted as an experimental artifact due to the injection of air during sampling. This air contamination relates to difficulties during sampling on board, i.e. difficulties

during squeezing of the individual liner segments, which led other liner segments to be exposed to air for an unusually long time, fostering the accumulation of some atmospheric air in the core.

The parameter A describes the amount of air per unit of water, and z^i describes the volume fraction of gas i in dry air (Kipfer et al., 2002). Finally, the term C^i_{terr} denotes a possible terrigenic component of gas i, which, in this study, is only present for helium.

The model of eq. (1) was used on the measured concentrations of He—Xe to perform a regression for the parameters A and C^i_{terr} (for details on the numerical methods, see Aeschbach-Hertig et al., 1999). The pressure was fixed at 1 atm and the water temperature and salinity were fixed at the in-situ values known in the Azores region for both sites (T = 3.5 °C, S = 35 ‰ at 2175 m (Southern Plateau) and T = 6 °C, S = 35.2 ‰ at 1225 m depth (Pico/Faial), see Amorim et al., 2017).

3. Results

The concentrations of the atmospheric noble gases (Ne, Ar, Kr, Xe) in the pore water relative to ASW concentrations are shown in Fig. 2 for the two sediment cores. At both sampling sites, the concentrations of Ar, Kr and Xe are close to the calculated concentrations of ASW in the Azores region. In both cores, there are samples which are supersaturated relative to the expected ASW concentrations which is, as mentioned in section 2, caused by air contamination during sampling. This air contamination is most evident for Ne concentrations, as Ne has the lowest solubility of the noble gases in Fig. 2 and is thus mostly affected by the addition of air (see, e.g., Kipfer et al., 2002). For the Southern Plateau, the largest air excess is observed in the samples from the middle of the core (between 1.5 m and 3.2 m) and is approximately 200% for neon. At the Faial/Pico site, three samples are subject to an air excess (1.03 m, 3.03 m and 4.63 m), with the highest excess of 800% for neon at the bottom of the core.

Some samples (most evidently the top sample at the Southern Plateau) have been subject to incomplete degassing during sample measurement, thus the measured concentrations (especially for Xe) are slightly lower than the calculated ASW concentrations. All noble gas concentrations for both sampling locations and the according errors are summarized in Table 1. As described in the methods sections, the concentrations of these gases were used to determine the atmospheric noble gas components in the sediment pore water and thus obtain the terrigenic helium component.

The terrigenic He concentrations are depicted in Fig. 3 for both sampling locations. The size of the errors is mostly attributed to the error propagation in the model regression (eq. (1)), and errors are particularly large for samples showing a large air excess.

At the Southern Plateau, the terrigenic helium concentration (Fig. 3a) is zero within the range of the error in the upper part of the core and increases slightly towards the bottom of the core. The terrigenic ^{3}He concentration increases with depth at both locations, however, the gradient is again more pronounced at Faial/Pico, which lies closer to the Terceira Rift and MAR. Our data show that at both sampling sites a terrigenic helium component is present in the sediment pore water, and that the component is more enriched closer to the Terceira Rift. Fig. 3 also indicates that no terrigenic He present at the top of the sediment core, close to the sediment-water interface at both locations.

The ^{3}He/^{4}He ratio of terrigenic component is shown in Fig. 4. The ^{3}He/^{4}He ratio increases with sediment depth at both locations, but the gradient is again stronger at Faial/Pico. The highest ^{3}He/^{4}He ratio, observed at the bottom of the Faial/Pico core, is about 9 R_A, and thus in the range of a typical MORB value. At both locations the ^{3}He/^{4}He ratio near the sediment/water interface is close to the atmospheric ratio. The He concentrations and the ^{3}He/^{4}He isotope ratio are summarized in Table 1.
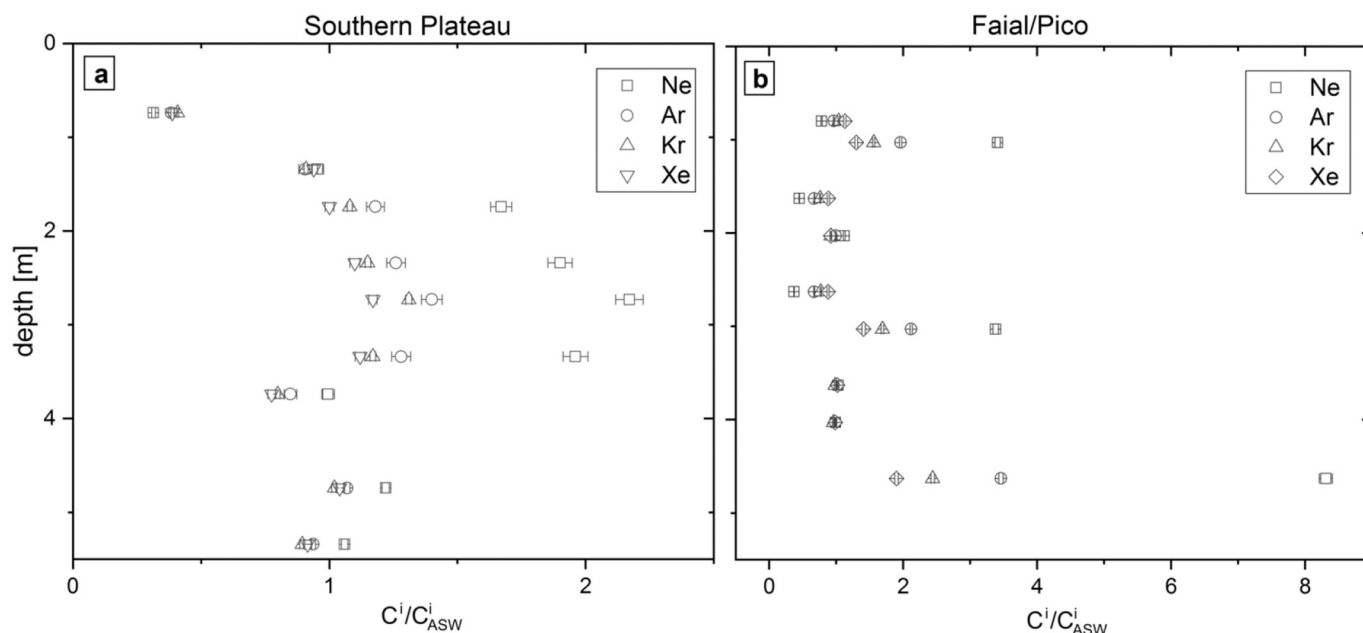
Fig. 2. Concentrations of Ne—Xe divided by the respective ASW concentrations, a) for Southern Plateau sampling location, b) for Faial/Pico sampling location. A value of 1 indicates the concentration of air-saturated ocean water, while a higher concentration indicates the presence of an atmospheric air component.

Table 1
Pore water concentrations of noble gases, helium isotope ratios and U and Th concentrations for both sampling locations with absolute errors. C_{ASW} denotes the expected concentrations (or the ^{3}He/^{4}He isotope ratio) of air saturated water calculated for in situ water temperature and salinity using the solubility data set recommended by Kipfer et al. (2000). U and Th concentrations were only measured at the top and bottom of the core.

Site	Depth [m]	Ne [10^{-7}] ccSTP/g	Ar [10^{-4}] ccSTP/g	Kr [10^{-8}] ccSTP/g	Xe [10^{-8}] ccSTP/g	He [10^{-8}] ccSTP/g	^{3}He/^{4}He [RA]	U [ppm]	Th [ppm]
Southern Plateau (M141–1186)	5.34	1.82 ± 0.03	3.27 ± 0.03	7.55 ± 0.09	1.14 ± 0.02	6.19 ± 0.04	2.3 ± 0.05	2.4 ± 1.0	4.9 ± 1.3
37°36.977′N	4.74	2.09 ± 0.03	3.73 ± 0.04	8.59 ± 0.10	1.29 ± 0.02	6.58 ± 0.05	1.94 ± 0.05		
27°32.522′W	3.74	1.71 ± 0.05	2.95 ± 0.09	6.75 ± 0.08	0.96 ± 0.02	5.65 ± 0.04	2.2 ± 0.05		
water depth 2175 m	3.34	3.36 ± 0.09	4.44 ± 0.13	9.86 ± 0.10	1.39 ± 0.03	10.2 ± 0.06	1.66 ± 0.04		
	2.73	3.72 ± 0.10	4.87 ± 0.15	11.0 ± 0.10	1.45 ± 0.03	10.2 ± 0.06	1.59 ± 0.03		
	2.34	3.25 ± 0.09	4.40 ± 0.13	9.68 ± 0.10	1.36 ± 0.03	8.59 ± 0.05	1.31 ± 0.04		
	1.74	2.86 ± 0.08	4.13 ± 0.12	9.11 ± 0.09	1.24 ± 0.03	8.85 ± 0.06	1.48 ± 0.03		
	1.34	1.63 ± 0.05	3.16 ± 0.10	7.68 ± 0.07	1.16 ± 0.02	4.33 ± 0.03	1.68 ± 0.05		
	0.74	0.54 ± 0.02	1.34 ± 0.04	3.45 ± 0.04	0.48 ± 0.01	1.21 ± 0.02	1.46 ± 0.06	3.9 ± 1.1	8.6 ± 2.0
C_{ASW}		**1.71**	**3.48**	**8.45**	**1.24**	**3.91**	**0.982**		
Pico/Faial (M141–1219)	4.63	13.9 ± 0.17	11.3 ± 0.11	19.1 ± 0.20	2.16 ± 0.04	65.7 ± 0.38	4.06 ± 0.03		
38°44,787′N	4.03	1.66 ± 0.02	3.22 ± 0.03	7.51 ± 0.08	1.12 ± 0.03	8.75 ± 0.06	4.82 ± 0.04	2.4 ± 0.3	3.2 ± 0.9
28°35,128′W	3.63	1.72 ± 0.03	3.31 ± 0.03	7.72 ± 0.10	1.16 ± 0.02	8.84 ± 0.06	5.03 ± 0.08		
water depth 1225 m	3.03	5.67 ± 0.07	6.95 ± 0.07	13.3 ± 0.16	1.60 ± 0.03	28.6 ± 0.17	4.51 ± 0.05		
	2.63	0.62 ± 0.01	2.20 ± 0.04	6.02 ± 0.07	1.00 ± 0.02	2.22 ± 0.03	3.86 ± 0.12		
	2.03	1.88 ± 0.03	3.26 ± 0.03	7.23 ± 0.09	1.04 ± 0.02	7.74 ± 0.05	4.14 ± 0.06		
	1.63	0.75 ± 0.01	2.21 ± 0.02	5.96 ± 0.07	1.00 ± 0.02	2.64 ± 0.03	3.03 ± 0.10		
	1.03	5.71 ± 0.07	6.43 ± 0.06	12.2 ± 0.13	1.47 ± 0.03	19.4 ± 0.12	2.42 ± 0.4		
	0.8	1.31 ± 0.02	3.17 ± 0.03	8.14 ± 0.09	1.28 ± 0.03	3.31 ± 0.03	1.07 ± 0.02	3.1 ± 0.50	4.2 ± 1.1
C_{ASW}		**1.68**	**3.28**	**7.83**	**1.14**	**3.87**	**0.982**		

4. Discussion

Our data show that a complete analysis of all noble gases in ocean sediments provide a useful tool to correct for possible sampling artefacts, such as air contamination, which then allows constraining the terrigenic He component in a robust and reliable manner.

At both sampling locations, an increasing ^{3}He/^{4}He gradient with depth and a the presence of terrigenic ^{3}He indicates that mantle helium is present in the sediment pore space, which is an indicator of hydrothermal fluids with a mantle-derived helium component. The observed gradient of terrigenic He in the upper 5 m of the sediment column leads to the conclusion that at these locations mantle He is migrating upward though the sediment body even though no vent structures or hydrothermally active sites are known in the vicinity.

Isotopically light mantle-type He becomes much more enriched with sediment depth at Faial/Pico than at the Southern Plateau (see Fig. 3b and Fig. 4). Thus, the enrichment of ^{3}He in fluids seems to intensify in the direction of the Mid-Atlantic Ridge and on the Terceira Rift. Due to
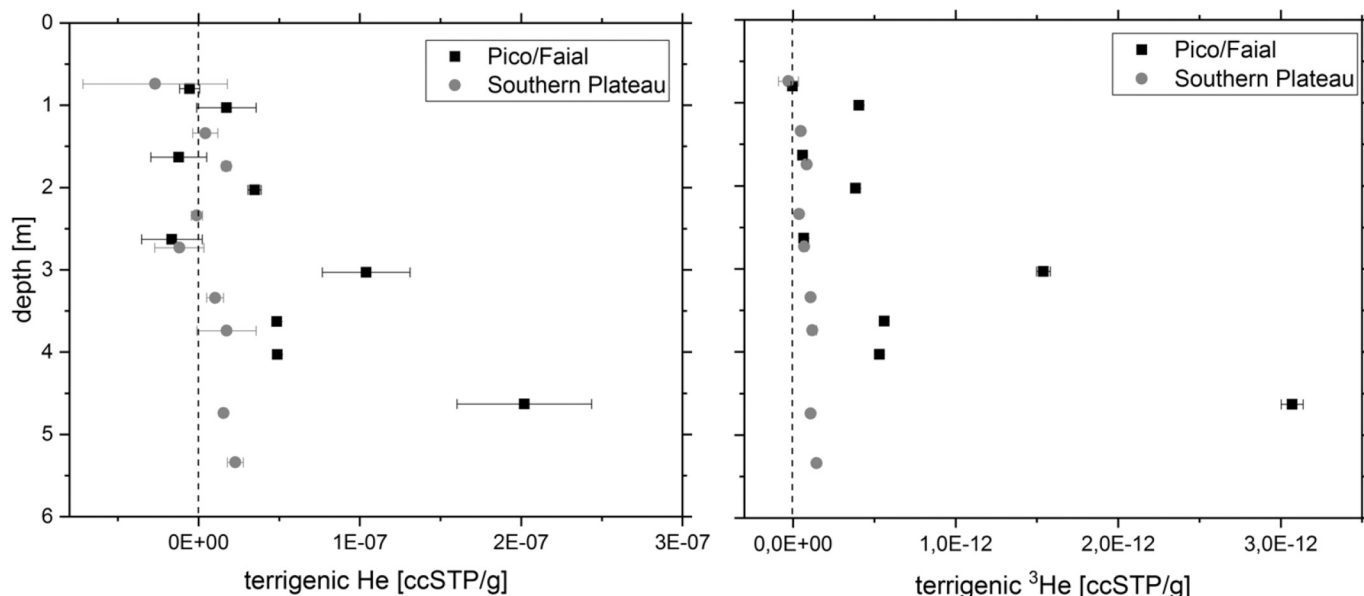
Fig. 3. Terrigenic concentrations for both sampling locations a) for total helium, b) for ^{3}He.
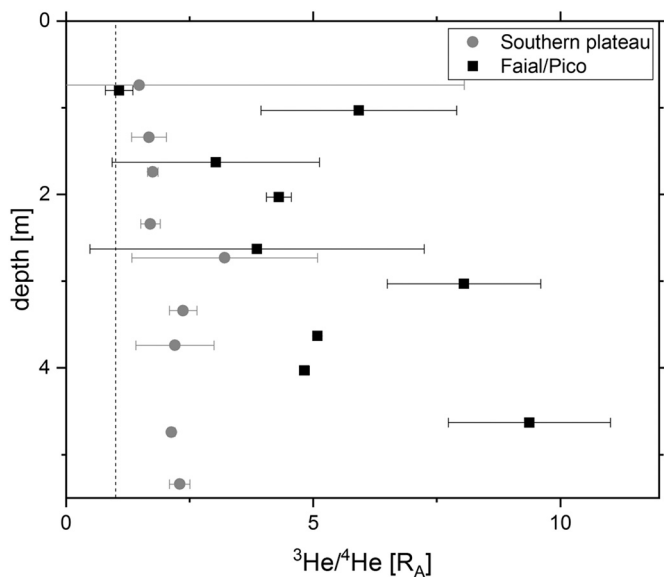

Fig. 4. ^{3}He/^{4}He ratio for both sampling locations in the Azores region. Data has been corrected for air excess, thus the plotted values represent ASW concentrations and the terrigenic (terrigenic) component. The dashed line represents a value of 1 R_A.

the gradients of terrigenic helium with sediment depth, we assume that at the two sampling sites the upward transport of mantle-derived solutes is mainly diffusion-controlled. If the transport properties of the sediments at both sampling locations are comparable, the stronger gradient implies a larger diffusive flux of ^{3}He at Faial/Pico. From this assumption it appears, that the presence of mantle fluids and hydrothermal activity in the Azores region is linked to the rift dynamics and the formation of oceanic crust at these two spreading centers.

The ^{3}He/^{4}He ratio in the sediments just below the sediment/water interface is very close the atmospheric ratio at both sampling sites (Fig. 4), and also terrigenic helium components in the pore water seem to vanish towards the upper part of the core. Thus, it can be argued that the local bottom water does not reflect the high ^{3}He/^{4}He ratio found in the sediments and cannot provide evidence for mantle helium

signatures. This leads to the assumption that evidence for hydrothermal activity might only be detectable in the sediment in the Azores region.

The fact that even at the Southern Plateau (about 100 km away from the Terceira Rift) a gradient of ^{3}He concentration with sediment depth is still observed (see Fig. 3), suggests that in the at the two sampling sites hydrothermal fluids migrate through the pelagic sediments over large areas of the ocean floor in a rather non-localized manner.

By plotting ^{3}He/^{4}He vs. Ne/He ratio in the sediments and fitting a line through the data, we can obtain the ^{3}He/^{4}He ratio of the source of the mantle-derived fluids at the Pico/Faial sampling location, as this ratio is represented by the y-axis section of the fit (Ne/^{4}He = 0, see Fig. 5) as the mantle can be assumed to be virtually Ne-free. The fitted line represents a mixing line between the ^{3}He-rich hydrothermal
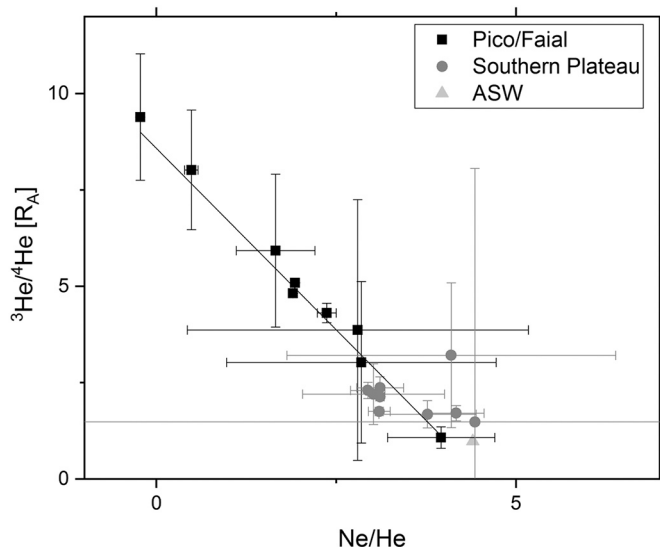

Fig. 5. ^{3}He/^{4}He ratio vs. Ne/He at both sampling locations. The plot represents mixing between ASW and the endmember feeding the hydrothermal system of the Azores. Data has been corrected for air excess, thus the plotted values represent ASW concentrations and a potential terrigenic component for all gases. A linear fit was applied to the data from Pico/Faial (black data points). Tracing back the fitted line line to Ne/He = 0 yields and estimate for the ^{3}He/^{4}He ratio of the original mantle source at this sampling location.

component and ocean bottom water (ASW). We obtain a ^{3}He/^{4}He ratio of about 8.6 ± 0.3 R$_A$ for noble gas data of Pico/Faial. This isotope ratio is characteristic for a MORB source, which we interpret to be the source of the fluids in the sediment pore water. Very similar ^{3}He/^{4}He ratios have been found in thermal waters and gas emissions on the islands of Pico and Faial (Jean-Baptiste et al., 2009).

We note that data in Fig. 5 have been corrected for air contamination during sampling, however, fitting the uncorrected values in the same manner yields a similar value of 8.1 ± 0.4 R$_A$ for the source of the hydrothermal fluids, thus the accuracy of the air correction does not play a major role for the determination of the helium isotope ratio of the terrigenic fluid source.

In Fig. 5, the data of the samples from the Southern Plateau scatter around the Pico/Faial mixing line but are much more similar to ASW values since these samples are less enriched in the mantle component. The lower ^{3}He/^{4}He ratios at this location are most likely the result of a lower flux of ^{3}He (see above) and, to a lesser extent, of the presence of radiogenic He in the sediments. Our U and Th data (Table 1) show that Th concentrations at the top and bottom of the core are considerably higher in the sediments of the Southern Plateau compared to the Pico/Faial site. Concentrations of U, however, are only slightly higher at the Southern Plateau towards the top of the core, and the same as at Faial/Pico at the bottom. The observation of an isotopically heavier He component as the consequence of radiogenic He accumulation at the Southern Plateau is in line with noble gas results from measurements in water and gas samples (Jean-Baptiste et al., 2009) on Sao Miguel.

Despite the fact that no vent structures or hydrothermally active sites are known in the vicinity, the ^{3}He/^{4}He ratios in the Azores region presented in this study are twice as high (maximum value of about 5 R$_A$ at Pico/Faial, see Table 1) as the isotope ratio found in a similar noble gas study in deep-sea sediments very close to a currently active black smoker system in the Pacific by Horstmann et al. (2021). The reason for the high ^{3}He/^{4}He ratios gradients observed in this study is most likely linked to the type of fluid transport. Our data suggest, that at the Azores hydrothermal fluids emanate over larger areas of the ocean floor through wide-spread pathways in the pelagic sediment body (non-localized transport). At the Nankai Trough off-shore of Japan diffusion-controlled He transport at the ocean floor has already been observed at sampling sites where a general ^{3}He enrichment was observed in the ocean bottom water, so only the analysis of He concentrations in the sediment pore water could allow the identification of fluid emission (Tomonaga et al., 2020). Additionally, the upward transport of hydrothermal fluids through the sediment body could be attenuated around the Azores, due to ash layers from volcanic eruptions in the ocean sediments, which have been observed during the *M141/1* cruise (Schmidt et al., 2020; Chang et al., 2021), and which might play a role in inhibiting a detectable input of hydrothermal fluids into the overlying ocean water. Such reduced transport through the sediment column in combination with the rather fast exchange of the bottom water around the Azores explain why hydrothermal fluids are not likely to accumulate in the local bottom water.

5. Conclusions

Our helium concentrations measured in pore water show that the ocean sediments around the Azores are enriched in isotopically light helium from a mantle source with a MORB-type helium signature and that the concentration of the terrigenic helium increases with depth. These data point to ongoing hydrothermal activity in the pelagic sediments in this region which allows ^{3}He-enriched fluids to emanate. Our data also indicate that in this region hydrothermal fluids migrate upwards through the pelagic sediment body in a non-localized manner over larger areas of the ocean floor. The results suggest that the diffusive flux of mantle He is stronger closer to the Terceira Rift and Mid-Atlantic Ridge, as concentration gradients of mantle helium with depth are steeper.

Our noble gas data mainly indicate diffusive transport of solutes

through the sediment column. However, these results are site-specific, therefore, the possibility of advective hydrothermal transport in the Azores region cannot be ruled out. Furthermore, we note that our results are only indicative for the uppermost meters of the sediment column and do not exclude the presence of advective transport along fissures and faults in the highly tectonized deeper part of the Azores Plateau. Investigations regarding this question are targeted by upcoming oceanographic expeditions by Geomar ('Azores Hot Vents' project).

Around the Azores, this mainly diffusion-driven transport mechanism results in a strong decrease of the mantle He signature in the sediment pore water towards the sediment/water interface, such that emanation of hydrothermal fluids can only be detected in the pore water of the sediments but not in the overlying open water column. This observation leads to the important conclusion that the absence of hydrothermal signals in ocean bottom water generally does not exclude the presence of hydrothermal fluids in the sediment column. Our findings thus highlight the importance of the analysis of noble gas concentrations in deep-sea sediments to trace hydrothermal activity, despite the experimental challenges and methodological effort of the respective analysis.

CRediT authorship contribution statement

E. Engelhardt: Investigation, Formal analysis, Writing – original draft, Visualization. **Y. Tomonaga:** Methodology, Formal analysis, Writing – review & editing. **M.S. Brennwald:** Conceptualization, Methodology, Writing – review & editing. **C. Maden:** Resources, Methodology, Software. **T.H. Hansteen:** Project administration, Funding acquisition, Resources, Writing – review & editing. **C. Schmidt:** Investigation, Writing – review & editing. **R. Kipfer:** Conceptualization, Supervision, Methodology, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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