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Helium transport in sediment pore fluids of the Congo-Angola margin

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Abstract:

During the ZaïRov2 cruise of the ZaïAngo project (1998–2000) on the passive Congo-Angola margin, several gravity cores were analyzed for helium isotopic composition of sedimentary pore waters in two cold fluid seepage zones: the Astrid slide area and the Regab giant pockmark. Gas concentration and isotopic composition are presented along with thermal data in terms of the origin and circulation of fluids. Helium isotope data lie on a mixing line between bottom seawater and an almost pure radiogenic. Helium and temperature vertical profiles are well described by the classic diffusion-advection equation. On the basis of He profiles, we estimate the advection rate on the rim of the pockmark between 1.2 and 2.3 mm/a. The He flux derived for a pure diffusive regime (2.4×10^{-8} mol/m²/a) can favorably be compared to literature data and contrasts with the flux computed close to the pockmark center (1.9×10^{-7} mol/m²/a). Helium depth profiles turned to be more sensitive to advection rate than temperature profiles are.

Keywords: helium; pockmark; pore fluids; Regab; sediment; Zaire-Angola margin.

22 **1. Introduction**

23

24 Continental margins are dynamic environments favouring the generation of both bio-
25 and thermogenic gases and fluid migration. Compaction and overpressure (like in oil
26 reservoir leaking, aquifer freshwater expulsion or magmatic intrusion) cause fluids to
27 migrate through the sedimentary column and reach the seafloor (Ballentine et al, 2002;
28 Berndt, 2005). The consequences are shallow gas accumulations, gas or water seeps,
29 pockmarks, mud volcanoes or natural gas hydrates deposits, often associated with cold seep
30 communities and methane-derived authigenic carbonates. Based on these features, several
31 authors presented an overview of fluid venting sites in marine sediments on a global scale
32 (e.g. Fleisher et al., 2001; Judd, 2003; Mazurenko and Soloviev, 2003). A large majority of
33 these sites are the result of focused fluid migration through sediments.

34 ZaiAngo (1998-2000), a joint project between IFREMER and the TOTAL oil company,
35 was dedicated to the geological and geochemical exploration of a large area of the Congo-
36 Angola margin (Savoie et al., 2000). The research project also included the study of cold
37 seeps and gas hydrate deposits associated with several previously identified pockmarks.
38 Pockmarks were first observed on sidescan sonar records off Nova Scotia (Canada) and
39 described by King and MacLean (1970) as “cone-shaped depressions possibly formed by
40 either ascending gas or subsurface water leakage from underlying sediments”. They usually
41 appear in fine-grained sediments as circular depressions ranging in size from small units
42 (1-10 m wide, <1 m deep) to larger structures ten to several hundred meter wide and up to
43 45 m deep (Hovland et al., 2002). Hovland and Judd (1988) observed that pockmarks occur
44 all over the world and in a wide variety of geological settings. As part of the ZaiAngo
45 project, the ZaiRov2 cruise (December 2000) onboard the RV L’Atalante was aimed at
46 exploring two areas of the Lower Congo Basin : the Astrid slide and the giant Regab

47 pockmark. Here, we focus on helium concentration and isotopic composition in interstitial
48 pore-fluids recovered from several gravity cores, and discuss the origin and circulation of
49 fluids.

50 The predominant source of ^4He in the Earth is from the radioactive decay of U and Th
51 whereas most of the ^3He is primordial in origin. Owing to their chemical inertness and
52 contrasted composition in the various earth reservoirs, helium isotopes have been used
53 extensively as tracers of mantle volatile inputs and provide information on fluid origin not
54 available from the study of active chemical species (Lupton, 1983). Precise measurements
55 of helium concentrations also allow us to quantify diffusive and advective fluxes through
56 the sediment column and to determine pore fluid advection rates (Barnes and Clarke, 1987;
57 Sayles and Jenkins, 1982). However, in spite of the potential of this geochemistry tool, its
58 use in pore-water studies has been hampered by the difficulties in collecting good samples
59 (i.e., without gas loss and/or contamination problems). Here we used a new sampling and
60 extraction method which allows the quantitative analysis of helium isotopes (Chaduteau et
61 al., 2007).

62

63 **2. Geological setting**

64

65 The passive continental margin of West Africa originates from the break-up of the
66 Gondwana supercontinent (Rabinowitz and Labrecque, 1979). The rifting of South America
67 and Africa and the opening of the South Atlantic ocean basin started at Early Cretaceous
68 (130 Ma). The Angola margin is a non-volcanic margin (Contrucci et al., 2004). From East
69 to West, reflection and refraction seismic data show four different domains (Fig. 1) : (1) a
70 domain with 30-km thick continental crust, (2) a domain where the crust thins from 30 km
71 to about 5 km, (3) a transitional domain with a crust not recognized as continental or

72 oceanic and (4) a domain of 6-km thick oceanic crust (Contrucci et al., 2004; Moulin et al.,
73 2005).

74 The sedimentary series of the Congo-Angola basin show three main units, which
75 correlate with three tectonic phases (Marton et al., 2000): pre-rift continental deposits
76 (Jurassic), syn-rift lacustrine deposits (early Cretaceous) and post-rift marine deposits
77 (Aptian to present). Following a large accumulation of evaporites during the Aptian, the
78 post-rift stratigraphy is characterized by an aggradation of a carbonate/siliclastic ramp from
79 Albian to Eocene, a truncation by a major erosional unconformity at Oligocene and the
80 progradation of a terrigenous wedge from Miocene to present (Séranne et al., 1992). This
81 switch in marine succession was initiated independently from any tectonic forcing and can
82 be explained by the transition from a greenhouse to an “ice-house” period. During the ice-
83 house period, high-frequency sea-level changes and an alternating drier and wetter climate
84 enhanced continental weathering (Séranne et al., 1999). The increased terrigenous input to
85 the margin has led to the formation of the Zaïre system, a large turbiditic submarine fan
86 directly fed by the Zaïre River and characterized by numerous turbiditic paleochannels. It
87 extends from the base of the slope (at about 2000 m depth) to the Angola abyssal plain with
88 a water depth of at least 5000 m, representing a total length of about 800 km (Droz et al.,
89 2003).

90 The location of the Regab and Astrid sites is shown in Figure 2. Regab is an active giant
91 pockmark, located at 3160 m depth in the abyssal domain less than 10 km north of the
92 channel system of the Zaïre fan. The large size of the pockmark, 800 m wide and 20 m deep
93 on average, results from the collapse of a cluster of several smaller pockmarks (Ondréas et
94 al., 2005). Gas hydrate outcrops are present on the seabed. A massive hydrate layer was
95 also observed at 12 m below the seafloor in one core (Charlou et al., 2004). Astrid is
96 located at 2820 m depth in the Zaïre deep-sea fan 80 km north of the channel system. It

97 corresponds to a gravity slide area where a cluster of pockmarks is observed. Whereas
98 Regab is in the oceanic domain, Astrid is in the transitional domain of the margin (Fig. 1).
99 According to the seismic velocity profiles of Contrucci et al. (2004) and Moulin and al.
100 (2005), Regab is on top of 6 km of sediment and 6 km of oceanic crust while Astrid is on
101 top of 9-10 km of sediment and 6 km of transitional crust.

102

103 **3. Sampling and methods**

104

105 3.1. Core locations

106

107 Four Küllenberg cores equipped with 9 thermistors out-rigged onto the piston corer
108 (Harmegnies and Landuré 2003) were taken for gas sampling : one in the Astrid area, KZR-
109 33 (13.65 m long), and three in the Regab area : KZR-37 (13.09 m long), KZR-38 (13.74 m
110 long), and KZR-40 (11.93 m long). The Astrid core is located on a gravity slide headscarp.
111 In the Regab area, KZR-37 and KZR-38 cores are located outside the pockmark,
112 respectively at 2 km and 1 km west of the centre, whereas KZR-40 stands inside the
113 pockmark at the western edge (Fig. 3). Core recovery at the centre of the pockmark was
114 impossible due to soft sediments falling through the core catcher.

115

116 3.2. Helium isotopes

117

118 Sampling was carried out immediately following the core retrieval and its cutting into
119 1 m long segments, and was done prior to any other core manipulation to minimize
120 potential gas loss and atmospheric contamination. The principle of the sampling technique
121 was to use copper tubes (1.2 cm OD, 25 cm in length) equipped with a small piston to take

122 mini-cores at both ends of each segment. The copper tube was tightly sealed with metallic
123 clamps. Back in the laboratory, each copper tube was placed on a vacuum line and sediment
124 was transferred into a glass bulb by applying pressurized degassed (helium-free) water at
125 one end of the tube. Helium was then extracted from the sediment slurry using a standard
126 method developed for water samples and analysed by mass spectrometry with a MAP 215-
127 50 spectrometer (Jean-Baptiste et al., 1992). The extraction blank is typically 1% of the
128 total helium signal. The 2-sigma uncertainty in the $^3\text{He}/^4\text{He}$ ratio is about 3%. For helium
129 concentrations, error bars are indicated in Table 1. Full details of the sampling method and
130 analytical procedure are available in Chaduteau et al., 2007.

131

132

133 3.3. Temperature profiles

134

135 Temperature profiles and heat flow determinations were obtained on all studied cores.
136 Temperature was measured with a single-penetration probe equipped with 9 thermistors.
137 Conductivities were measured onboard the ship using a needle probe technique (Von
138 Herzen and Maxwell, 1959) with a typical spacing of ~ 20 cm. Heat flow was determined
139 as the product of thermal conductivity and temperature gradient following the Bullard
140 method (Harmegnies and Landuré, 2003).

141

142 3.4. Dating of sediments

143

144 KZR-33, KZR-37 and KZR-40 cores were run through an Avaatech X-ray fluorescence
145 (XRF) Core Scanner (Richter et al., 2006) for Ca analysis. Although results are semi-
146 quantitative, this non-destructive technique provides rapid high resolution records of the

147 relative variability in elemental composition. An age model was then established from the
148 radiocarbon dating of handpicked foraminifera from Ca-rich levels and from the correlation
149 of the CaCO₃ profiles of the different cores. KRZ-38 was not available for analysis at the
150 time of our study. Therefore, KZR-38 age model was assumed to be identical to nearby
151 KZR-37. AMS radiocarbon analyses were performed at the LMC14 facility in Saclay
152 (France).

153

154 **4. Results**

155

156 4.1. Helium concentration and isotopic ratio

157

158 Helium isotope results are displayed in Table 1, along with ²⁰Ne concentrations. Helium
159 concentrations range between 5.51 and 52.86 × 10⁻⁸ ccSTP/g of porewater (*i.e.* between
160 2.46 × 10⁻¹² and 2.36 × 10⁻¹¹ mol/g of porewater), with clearly radiogenic ³He/⁴He ratios
161 between R= 0.09 and R= 0.79 Ra (where R is the ³He/⁴He ratio of the sample and Ra the
162 atmospheric ratio, Ra= 1.38×10⁻⁶). The average composition of bottom waters (subscript
163 'bw') in the study area is available from the measurements made by the University of
164 Bremen (<http://whpo.ucsd.edu/>) at two nearby WOCE (World Ocean Circulation
165 Experiment) stations, A13-205 and A13-213: ⁴He_{bw} = 4.12 × 10⁻⁸ccSTP/g (1.84 ×
166 10⁻¹²mol/g), (³He/⁴He)_{bw}=1.04 Ra, ²⁰Ne_{bw} = 1.62 × 10⁻⁷ccSTP/g (7.23 × 10⁻¹²mol/g). In
167 pore-waters, neon concentrations in excess of the bottom water value are indicative of a
168 slight atmospheric contamination (most likely due to tiny air bubble entrapment at the
169 surface of the copper tube during sampling) and can be used to correct the ⁴He and ³He
170 results using the ⁴He/²⁰Ne air ratio (0.3185) and the atmospheric ³He/⁴He ratio:

171

172
$${}^4\text{He}_{\text{corrected}} = {}^4\text{He}_{\text{measured}} - ({}^{20}\text{Ne}_{\text{measured}} - {}^{20}\text{Ne}_{\text{bw}}) \times ({}^4\text{He}/{}^{20}\text{Ne})_{\text{air}}$$

173

174
$${}^3\text{He}_{\text{corrected}} = {}^3\text{He}_{\text{measured}} - ({}^{20}\text{Ne}_{\text{measured}} - {}^{20}\text{Ne}_{\text{bw}}) \times ({}^4\text{He}/{}^{20}\text{Ne})_{\text{air}} \times ({}^3\text{He}/{}^4\text{He})_{\text{air}}$$

175

176 For most samples, the correction made to ${}^4\text{He}$ concentrations is small (between 0.5 and
177 2% - see Table 1). ${}^3\text{He}$ is more sensitive to any added air component (see Table 1) since
178 ${}^3\text{He}$ concentrations, contrary to ${}^4\text{He}$, remain close to the oceanic bottom water background
179 throughout the sediment column.

180 Pore-water ${}^4\text{He}$ concentrations increase steadily with depth (Fig. 4). The slope of the ${}^4\text{He}$
181 profile becomes steeper as the distance from the centre of the pockmark increases. This
182 points to a flux of helium from below, which tends to decrease away from the pockmark
183 (see discussion below).

184

185 4.2. Temperature profiles - heat flow

186

187 Temperature versus penetration depth is plotted in Fig. 5. The temperature profiles for
188 the Regab cores (KZR-37, KZR-38 and KZR-40) are almost linear, apparently suggesting a
189 heat transfer dominated by conduction. The corresponding heat flows are 41.8, 46.3 and
190 45.2 mWm^{-2} respectively. For the Astrid core (KZR-33) the heat flow is higher, reaching
191 58.7 mWm^{-2} . Note that the temperature profile is shifted upward due to the shallower depth
192 of this core. These results are consistent with Lucazeau et al. (2004) who compiled a large
193 amount of heat flow data from the lower Congo basin. At a small scale, they observe
194 substantial heat flow variations in connection with salt diapirs but no temperature anomaly
195 related to fluid venting in active pockmarks could be detected. We will see in section 5.2
196 that this lack of any detectable thermal anomaly is consistent with the magnitude of the

197 upward advection rates deduced from the helium vertical profiles. At the scale of the
198 margin, the thermal trend between the oceanic domain ($42 \pm 3 \text{ mWm}^{-2}$), the transitional
199 domain ($52 \pm 10 \text{ mWm}^{-2}$) and the continental domain ($65 \pm 15 \text{ mWm}^{-2}$) - Lucazeau et al.,
200 2004 - is consistent with our own measurements for Astrid and Regab located in the
201 transitional and oceanic domains respectively. These regional variations can be explained
202 by a combination of in-situ heat production in the crust and mantle heat flow : the heat
203 production decreases offshore and is negligible in the oceanic domain whereas mantle heat
204 flow increases at the continent edge (Guillou-Frottier and Jaupart, 1995; Lucazeau et al.,
205 2004).

206

207 4.3. Age model - accumulation rates

208

209 Knowledge of the sedimentation rate is a key point in order to model fluxes. Calcium
210 records obtained by XRF are presented in Fig. 6a along with calibrated ^{14}C ages of the Ca
211 peaks. Comparison of the KZR-33 and KZR-37 profiles suggests that about 1 meter of
212 sediment is missing on top of core KZR-33, likely due to some landslide. Conversion of
213 radiocarbon ages to calendar ages was done using Intcal04 (Hughen et al., 2004). Beyond
214 26 cal ka BP, we relied on the new marine-derived ^{14}C calibration of Hughen et al. (2006)
215 which extends the calibration to ~ 50 ka. The age results versus depth are plotted in Fig. 6b.
216 Taking into account the one meter that has been lost on top of KZR-33, sedimentation
217 appears to be homogeneous over the whole area. The Holocene ($13.5 \pm 1.5 \text{ cm/ka}$) and
218 Glacial ($17 \pm 2.5 \text{ cm/ka}$) accumulation rates are within the uncertainty of each other, and we
219 conclude that the accumulation rate is approximately $16 \pm 4 \text{ cm/ka}$ over the whole period
220 studied. This accumulation rate is typical of the entire period back to early Pliocene (~ 5
221 Ma) during which about 700 m of sediment were deposited in this area (Gay et al., 2006a).

222

223

224 **5. Discussion**

225

226 5.1. Origin of helium

227 Helium isotope results are plotted in Fig. 7 using a R/Ra vs ($1/{}^4\text{He}$) mixing diagram
228 which allows to define the ${}^3\text{He}/{}^4\text{He}$ ratio of the helium in excess of the solubility
229 equilibrium. For comparison, we include the few data on sediment pore-waters published in
230 the literature. All our samples fall on a simple mixing line between a bottom sea water end-
231 member ($\text{R/Ra} \sim 1$; ${}^4\text{He}_{\text{bw}}/{}^4\text{He} = 1$) and an almost pure radiogenic ${}^4\text{He}$ source ($\text{R/Ra} =$
232 0.04). No trend is observed towards the MORB end-member ($\text{R/Ra} \sim 8$), as it is seen for
233 instance in the sediment-rich hydrothermal system of the Escabana Trough. This indicates
234 that the contribution from MORB ${}^3\text{He}$ is minimal in this system and that the helium excess
235 is overwhelmingly derived from U/Th radioactive decay in the underlying crust and
236 sedimentary column. As shown in Figure 7, this result is comparable to what is observed in
237 the Nankai Trough and the Japan Trench by Sano and Wakita (1985).

238

239 5.2. Fluid circulation and advection rates

240

241 Two-dimensional high resolution seismic profiles across the pockmark reveal a 300 m
242 chimney-like feature (Fig. 8) interpreted as an ascending movement of fluids (Ondreas et
243 al., 2005). On the lower slope of the Congo basin, Gay et al. (2006a) noted that a sinuous
244 belt of pockmarks mimics the meanders of a buried paleo-channel which could act as a
245 drain for interstitial fluids. In the Regab pockmark indeed, the chimney imaged by the
246 seismic data branches on an ancient buried channel levee system (Gay et al., 2006b).

247 Therefore, it seems likely that the Regab fluids originate from a shallow (~300 m) silty-
248 sandy paleo-channel reservoir from which they are expelled by the overpressure due to
249 compaction.

250 The helium vertical profiles for KZR-37, KZR-38 and KZR-40 are consistent with this
251 scheme : the three cores are close to each other, so their different slopes cannot be
252 explained by variations in sediment thickness, ⁴He diffusion or production rate. We have
253 also shown that sediment accumulation rates are similar for the three cores. Therefore, from
254 KZR-37 to KZR-40, the decreasing slopes reflect an increasing fluid upward circulation
255 when approaching the centre of the pockmark. This upward velocity can be estimated by
256 solving the helium diffusion/advection mass balance in the sediment column. To do this,
257 we first solved the helium diffusion/advection equation numerically in the transient mode
258 (i.e., with a growing sediment column simulating the sedimentation process) and compared
259 the results a simple steady-state solution which neglects sediment accumulation. The results
260 are identical, indicating that the sediment accumulation rate (~16 cm/ka) is not high enough
261 to play a significant role in the diffusion/advection process. Hence, the profiles can be
262 safely described by the classic steady-state diffusion/advection equation (Eq. 1; Fick, 1855)
263 for which an analytical solution is available :

264
$$D \frac{\partial C(x)}{\partial x} + C(x) \times V = J_0 \quad (\text{Eq. 1})$$

265 where x is depth ($x = 0$ at the seawater/sediment interface), C the helium concentration per
266 volume unit of the bulk sediment (concentration in the bulk sediment equals that in pore
267 water multiplied by the porosity n expressed as the volume of pore space per volume unit of
268 bulk sediment), D the He bulk diffusion coefficient, and J_0 is the helium flux through the
269 sediment column. With this formalism, the advection velocity V is defined by $V = v/n$
270 where v is the volume of water advected by unit of surface area and unit of time and n is the
271 porosity of the sediment ($n=0.86\pm 0.03$ from on-board measurements with Multi Sensor

272 Core Logger). Two boundary conditions are necessary to solve this equation analytically :
 273 the first condition is that $C(0)/n$ is equal to the He concentration measured in bottom
 274 seawater. For the second condition, based on the existence of a fluid reservoir (high
 275 permeability layer) at $x = 300\text{m}$ revealed by seismic imaging (Gay et al. 2006b), we choose
 276 to set a constant ^4He concentration (C_{deep}) in this paleo-channel reservoir at this depth,
 277 further called $H (= 300\text{m})$.

278 The helium concentration as a function of depth inferred from equation 1 is thus the
 279 following :

$$280 \quad \frac{C(x)}{C(0)} = \frac{\left(\frac{C_{deep}}{C(0)} - e^{-\frac{v}{D}H}\right)}{\left(1 - e^{-\frac{v}{D}H}\right)} \left(1 - e^{-\frac{v}{D}x}\right) + e^{-\frac{v}{D}x}$$

281 (2)

282

283 This analytical solution requires a constant porosity and tortuosity. In the studied zone,
 284 the variation of these parameters with depth is unknown. To estimate this effect, we ran a
 285 finite-difference numerical simulation with conservative estimates for these parameters
 286 (Briggs et al. 1998; Bahr et al. 2001). A comparison of the results indicates that the
 287 analytical solution may overestimate the calculated advection rate by 15% at most.

288 To determine the He concentration in the paleo-channel reservoir at $H = 300\text{ m}$ (C_{deep}),
 289 we assume a negligible advection for the KZR37 core, which leads to a concentration at
 290 depth of 2.1×10^{-6} ccSTP (9.4×10^{-11} mol) per cm^3 of bulk sediment. The best fit with the
 291 KZR40 helium profile is then obtained for a V/D value of $2.6 \times 10^{-2} \text{ m}^{-1}$ (see Fig. 7). The
 292 effective diffusion coefficient D is lower than the diffusion coefficient in free water ($D_0 \sim$
 293 $5 \times 10^{-9} \text{ m}^2/\text{s}$ - Jähne et al. 1987) due to the tortuosity of the bulk sediment. Traditionally, D
 294 is linked to D_0 by the relation $D = D_0 / (nF)$, where F is the "formation factor", in the range

295 1.3 to 3 for deep-sea sediments (Boyce, 1967). In diffusion experiments on several deep-sea
296 sediments, Ohsumi and Horibe (1984) found typical effective diffusion coefficients for
297 helium in superficial deep-sea sediments between 2 and $3 \times 10^{-9} \text{ m}^2/\text{s}$, in good agreement
298 with the formation factors determined by resistivity measurements. Thus we adopt a D
299 value of $(2.5 \pm 0.5) \times 10^{-9} \text{ m}^2/\text{s}$. It follows that advection v in the KZR40 core is in the range
300 1.2-2.3 mm/a. The modelled curves are included in Fig. 9.

301 The He flux in the pure diffusive regime is deduced from KZR37 profile : $2.4 \times$
302 $10^{-8} \text{ mol/m}^2/\text{a}$. This value supports the oceanic crustal flux estimated by Torgersen (1989)
303 from the production rate: he obtained a lower limit of $1.73 \times 10^{-8} \text{ mol/m}^2/\text{a}$ (considering
304 400m of 80% carbonated sediment on top of the crust) and an upper limit of $9.4 \times$
305 $10^{-8} \text{ mol/m}^2/\text{a}$. It is also consistent with the few data derived from sediment
306 measurements we found in the literature: $[0.6-3.3] \times 10^{-8} \text{ mol/m}^2/\text{a}$ (Barnes and Bieri, 1976)
307 and $[0.1-2.1] \times 10^{-8} \text{ mol/m}^2/\text{a}$ (Sano and Wakita, 1987). More recently Well et al. (2001)
308 derived an average flux of $(5.2 \pm 2) \times 10^{-8} \text{ mol/m}^2/\text{a}$ from the helium excess in Pacific deep
309 waters.

310 For KZR40, the best estimate of V/D ($2.6 \times 10^{-2} \text{ m}^{-1}$) corresponds to a flux of 1.9×10^{-7}
311 $\text{mol/m}^2/\text{a}$. This value is an order of magnitude higher than that of the pure diffusive regime,
312 illustrating the focusing effect of the pockmark. In comparison, the ^4He in-situ production
313 can be estimated to $1.2 \times 10^{-9} \text{ mol/m}^2/\text{a}$ using a typical crustal composition for the mineral
314 fraction of the sediment ($U=1 \text{ ppm}$ and $\text{Th}/U=4$), and is in fact negligible.

315

316 An advection rate of the order of a few mm/a is not in contradiction with the temperature
317 data, even if at first sight the similarity of the temperature profiles between KZR-37, KZR-
318 38 and KZR-40 would suggest a purely conductive heat transfer for all the cores. In fact the
319 same type of diffusion/advection equation can be solved for heat. The simulated

320 temperature profiles for different advection rates are plotted in Fig. 10. It shows that
321 temperature profiles are far less sensitive to advection than He profiles and that an increase
322 of advection between KZR-37 and KZR-40 is not inconsistent despite similarities between
323 the two temperature profiles. This is due to the thermal diffusion coefficient ($\sim 1.7 \times 10^{-7}$
324 m^2/s) being almost two orders of magnitude higher than the He diffusion coefficient.
325 Hence, helium is clearly a more sensitive tracer of fluid movements in the sediment
326 column.

327

328 The KZR-33 helium profile, located in the Astrid zone, lies in an intermediate position
329 (Fig. 7). Whether this profile is influenced by advection (since the Astrid site is a cluster of
330 pockmarks) or by a higher flux linked to the greater heat flux from the continental crust due
331 to its geographical position closer to the continental domain is difficult to say. As a matter
332 of fact we lack the appropriate geophysical data to set realistic lower boundary conditions
333 that would allow us to test both hypothesis.

334

335

336 **6. Conclusions**

337

338 We measured helium and methane vertical concentration profiles in sediment pore-
339 waters from two cold fluid seepage zones of the Congo-Angola margin : the Astrid slide
340 area and the Regab giant pockmark. For helium, we used a new method for gas sampling
341 and recovery which avoids the long-standing gas loss and/or air contamination problems
342 classically associated with pore-water sampling. The main results of the present study are
343 the following :

344 - Sedimentation is homogeneous over the whole Regab area, with an accumulation rate
345 of 16 ± 4 cm/ka.

346 - In the Astrid area, heat flow (58.7 mWm^{-2}) is slightly larger than in the Regab zone
347 ($44.4 \pm 2.3 \text{ mWm}^{-2}$) in agreement with their geographical position with respect to the
348 transition between the continental and oceanic domains.

349 - Helium isotopes data lie on a mixing line between bottom seawater and a radiogenic,
350 showing that the helium signature is overprinted by the ^4He production from U/Th
351 radioactive decay in the sediment column and underlying crust.

352 - Helium and temperature vertical profiles are well described by the classic steady-state
353 diffusion-advection equation. In the Regab zone where three different cores could be
354 compared, helium profiles show increasing advection rates towards the centre of the
355 pockmark For core KZR40 located inside the pockmark, we calculate an advection rate
356 in the range 1.2-2.3 mm/a.

357 - The derived He flux is $2.4 \times 10^{-8} \text{ mol/m}^2/\text{a}$ for the pure diffusive regime (KZR37) wich
358 is in good agreement with both estimations computed from He production rate
359 (Torgersen, 1989) and from experimental measurements (Barnes and Bieri, 1976; Sano
360 and Wakita, 1987, Well et al, 2001). For the core closest to the pockmark center
361 (KZR40), the flux is estimated to $1.9 \times 10^{-7} \text{ mol/m}^2/\text{a}$, an order of magnitude higher than
362 for KZR37, illustrating the focusing effect of the pockmark.

363 - Comparison between helium and thermal profiles shows that helium is a far more
364 sensitive tracer of water movements than temperature.

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366

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375 **References**

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506

507 **Figure captions**

508

509 Fig. 1 : a/ Major structural domains of the Congo-Angola margin based on seismic and
510 gravity data : zone I is the unthinned continental domain; zone II, the domain where the
511 crust thins; zone III, the transitional domain and zone IV, the oceanic domain (map adapted
512 from Moulin et al., 2005). b/ schematic geological cross-section of South-Atlantic margin
513 offshore Congo and Angola (taken from Huc, 2004)

514

515 Figure 2 : Location of Astrid and Regab areas on the bathymetric map of the Congo-Angola
516 margin.

517

518 Figure 3 : Location of the Regab zone on a Simrad EM-12 multibeam sonar image of the
519 Zaire deep-sea fan.

520

521 Figure 4 : ^{20}Ne -corrected pore-fluid helium isotope data (^4He per gram of pore water and
522 R/Ra). When not visible, error bars are smaller than the symbols.

523

524 Figure 5 : In-situ temperature profiles and thermal data. Note that the KZR-33 profile
525 (Astrid zone) is shifted upward due to shallower depth.

526

527 Figure 6 : a/ KZR-33, KZR-37 and KZR-40 cores XRF calcium profiles. Figures are
528 calendar ages (cal year BP) deduced from radiocarbon dating. b/ Age model for KZR-33,
529 KZR-37 and KZR-40.

530

531 Fig. 7 : (R/Ra) vs $({}^4\text{He}_{\text{BSW}}/{}^4\text{He})$ for measured sediment pore-fluids. Data from the Nankai
532 Trough and the Japan Trench (Sano and Wakita, 1985), from the Ontong-Java plateau and
533 Chatham Rise (Wakita et al, 1985), from the Escabana Trough (Ishibashi et al., 2002) and
534 from different ODP sites in the Pacific and the Atlantic (Barnes and Clarke, 1987) are
535 shown for comparison. For Sano and Wakita (1985) and Wakita et al (1985) for which only
536 ${}^{20}\text{Ne}/{}^4\text{He}$ ratios are available, we use the neon solubility data of Weiss (1971). The two
537 lines represent mixing trends between bottom sea-water ($R \approx Ra$) and MORB ($R \approx 8Ra$) or
538 pure radiogenic helium sources respectively.

539

540 Figure 8 : Two dimensional high resolution seismic profile across the Regab pockmark : the
541 seismic chimneys are interpreted as an ascending movement of fluids from an ancient
542 buried channel-levee system (after Gay et al., 2006b)

543

544 Fig. 9 : Neon-corrected ${}^4\text{He}$ concentrations in sediment pore-fluids. Error bars are given in
545 Table 1 (they are of the size of the symbols for KZR37 and 38 and slightly larger for
546 KZR 40). Solid curves correspond to the analytical solution of the steady-state advection-
547 diffusion equation (see text) for various V/D values (see figures on the curves). For $D=$
548 $2.5 \times 10^{-9} \text{ m}^2/\text{s}$ (see text), the purple curve corresponds to a 10 mm/a advection, the solid red
549 curve to 1.8 mm/a (the dashed red curves to 1.45 and 2.01 mm/a respectively), and the grey
550 curve to 0.1 mm/a.

551

552 Figure 10 : Comparison between measured and modelled temperature profiles using the
553 same type of boundary conditions as for helium.

554

Sample no.	Depth (m)	⁴ He corr (10 ⁻⁸ cc/g)	σ (10 ⁻⁸ cc/g)	correction (%)	³ He corr (10 ⁻¹⁴ cc/g)	σ (10 ⁻¹⁴ cc/g)	correction (%)	²⁰ Ne (10 ⁻⁷ cc/g)	R/Ra corr
KZR33- Astrid									
3-4	3.99	14.17	0.27	0.6	6.15	0.24	2.00	1.65	0.31
5-6	5.99	17.88	0.29	-	6.17	0.13	-	1.56	0.25
7-8	8.01	20.62	0.49	22.5	6.38	0.44	56.50	3.49	0.22
9-10	10.01	28.33	0.49	1.0	5.86	0.25	6.60	1.71	0.15
KZR37- Regab 2km W									
S-1	0.91	5.51	0.16	0.1	6.03	0.23	0.10	1.62	0.79
2-3	2.73	6.57	0.11	-	6.68	0.15	-	1.55	0.74
3-4	3.91	7.30	0.18	2.0	6.17	0.23	3.20	1.66	0.61
4-5	4.89	8.10	0.14	-	6.46	0.14	-	1.61	0.58
5-6	5.89	8.25	0.20	5.5	5.65	0.25	10.50	1.77	0.50
6-7	6.89	9.22	0.21	0.4	6.37	0.24	0.80	1.63	0.50
7-8	7.89	9.72	0.16	-	6.27	0.14	-	1.58	0.47
8-9	8.89	10.19	0.22	1.4	6.15	0.25	3.20	1.66	0.44
9-10	9.58	11.16	0.24	1.7	5.94	0.25	4.40	1.68	0.39
11-12	11.58	13.35	0.27	3.4	5.87	0.24	10.10	1.77	0.32
12-13	12.58	14.41	0.24	-	6.21	0.14	-	1.61	0.31
KZR38- Regab 1km W									
1-2	2	6.83	0.19	6.5	6.08	0.25	9.80	1.77	0.64
2-3	3	8.22	0.25	0.6	6.15	0.21	1.10	1.63	0.54
4-5	5	9.28	0.22	6.6	5.32	0.23	14.60	1.83	0.42
5-6	6	10.53	0.18	-	6.43	0.15	-	-	0.44
7-8	8	12.15	0.25	1.2	6.38	0.20	3.10	1.67	0.38
8-9	9	12.72	0.26	2.4	6.15	0.20	6.60	1.72	0.35
9-10	10	14.16	0.28	1.4	6.25	0.19	4.20	1.68	0.32
10-11	11	15.44	0.29	0.8	6.36	0.20	2.60	1.66	0.30
11-12	12	17.72	0.45	-	6.74	0.15	-	1.57	0.28
KZR40- Regab inside									
1-2	1.78	9.04	0.15	-	6.15	0.13	-	1.59	0.49
2-3	2.63	15.68	0.30	1.1	6.08	0.24	3.90	1.67	0.28
3-4	3.59	24.02	0.43	1.5	6.42	0.24	7.20	1.73	0.19
5-6	5.42	34.22	0.59	1.2	6.54	0.26	8.00	1.75	0.14
7-8	7.19	44.58	0.77	0.1	6.75	0.24	1.30	1.64	0.11
8-9	8.07	52.86	0.91	1.9	6.30	0.27	18.10	1.93	0.09
WOCE	-	4.12			5.90			1.62	1.03

Ra is the atmospheric ratio (1.38x 10⁻⁶)

Table 1 : ²⁰Ne-corrected pore-fluid helium isotope data (concentrations are per gram of pore water)

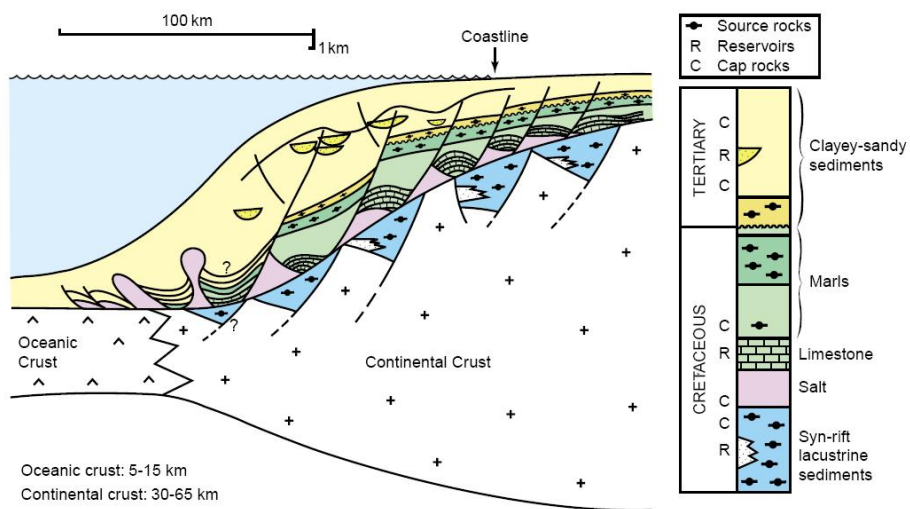
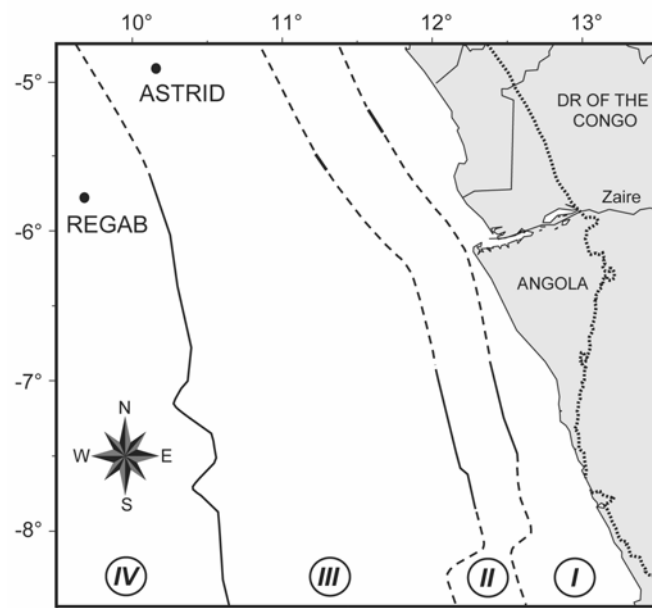


Fig. 1

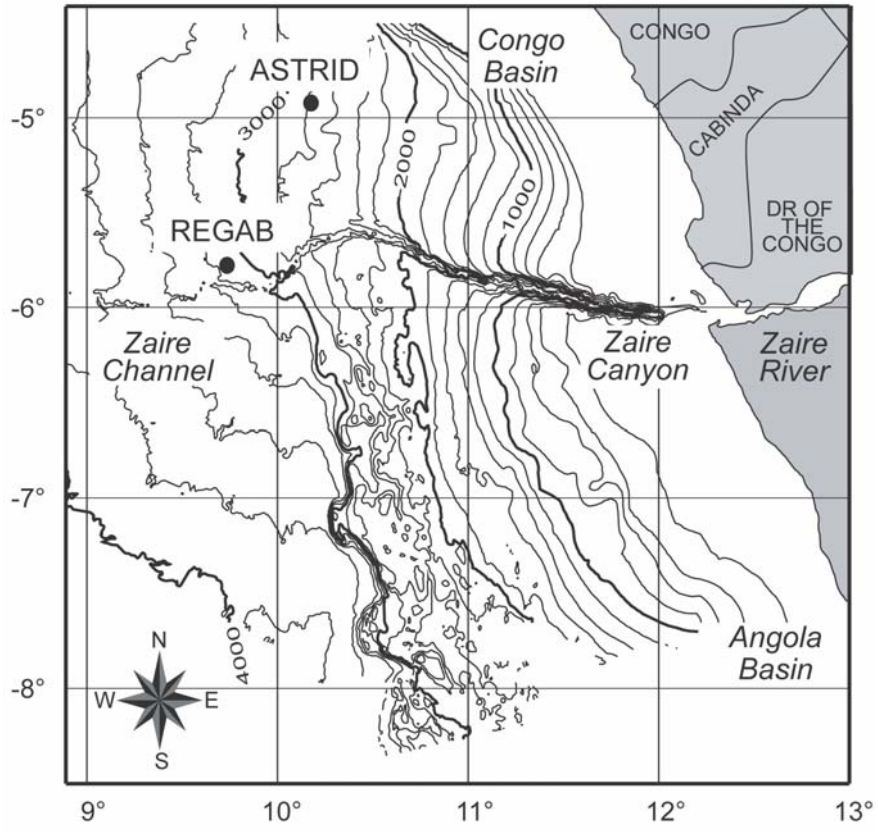


Fig. 2

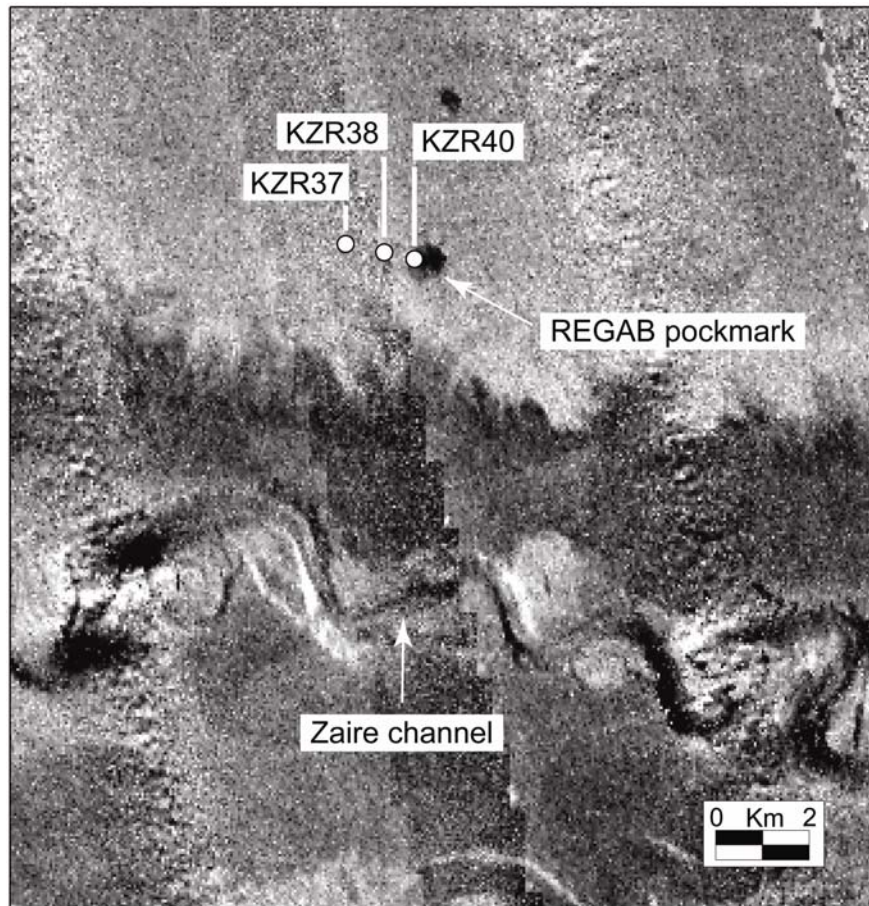


Fig. 3

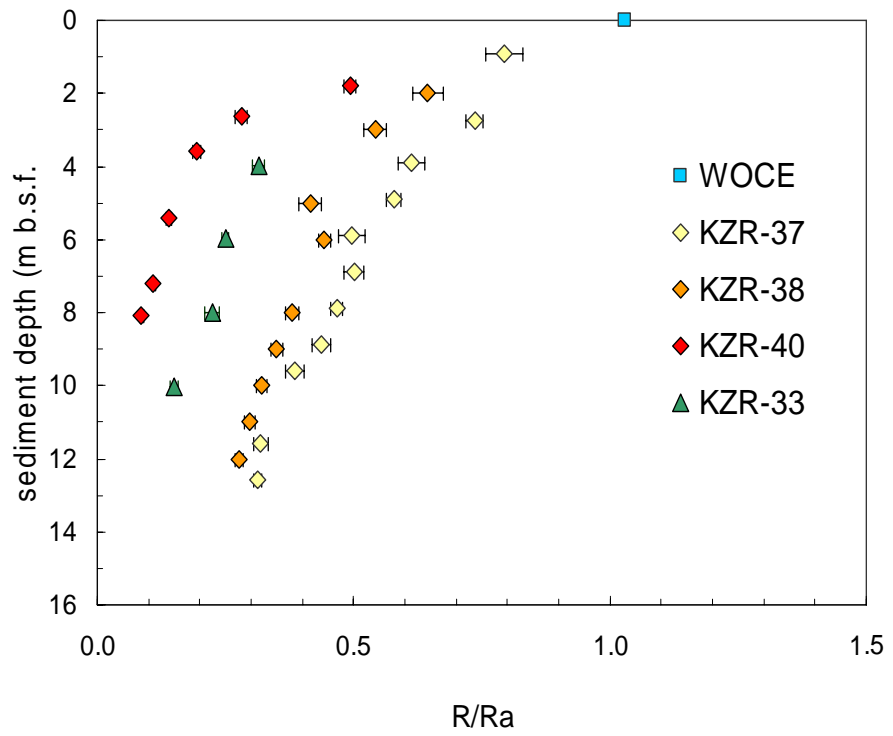
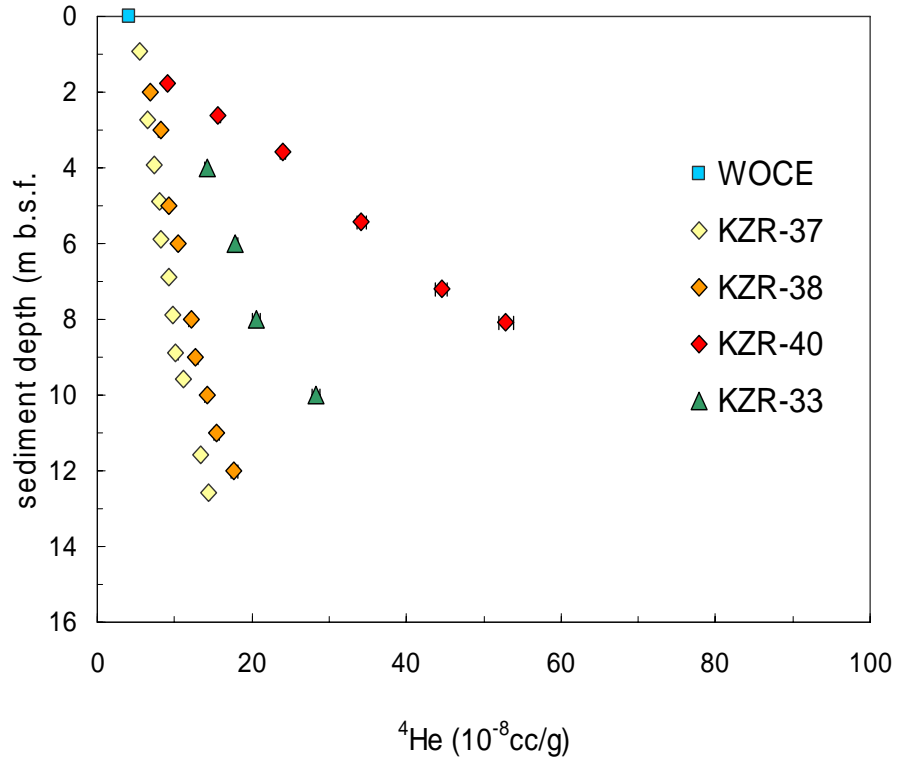


Fig. 4

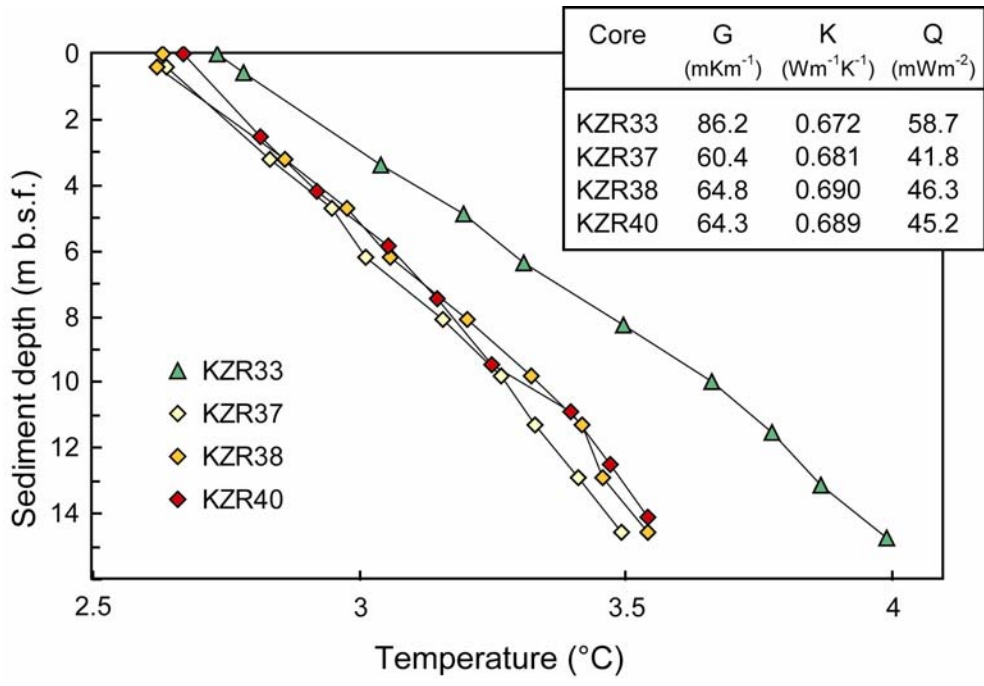


Fig.5

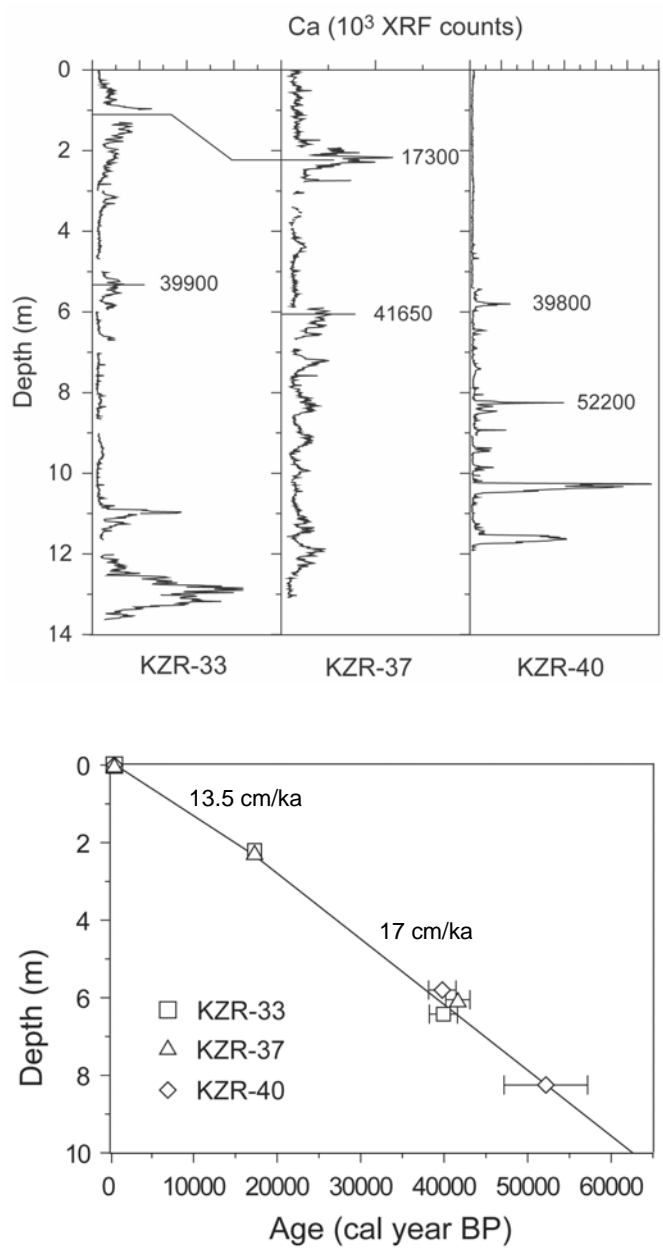


Fig. 6

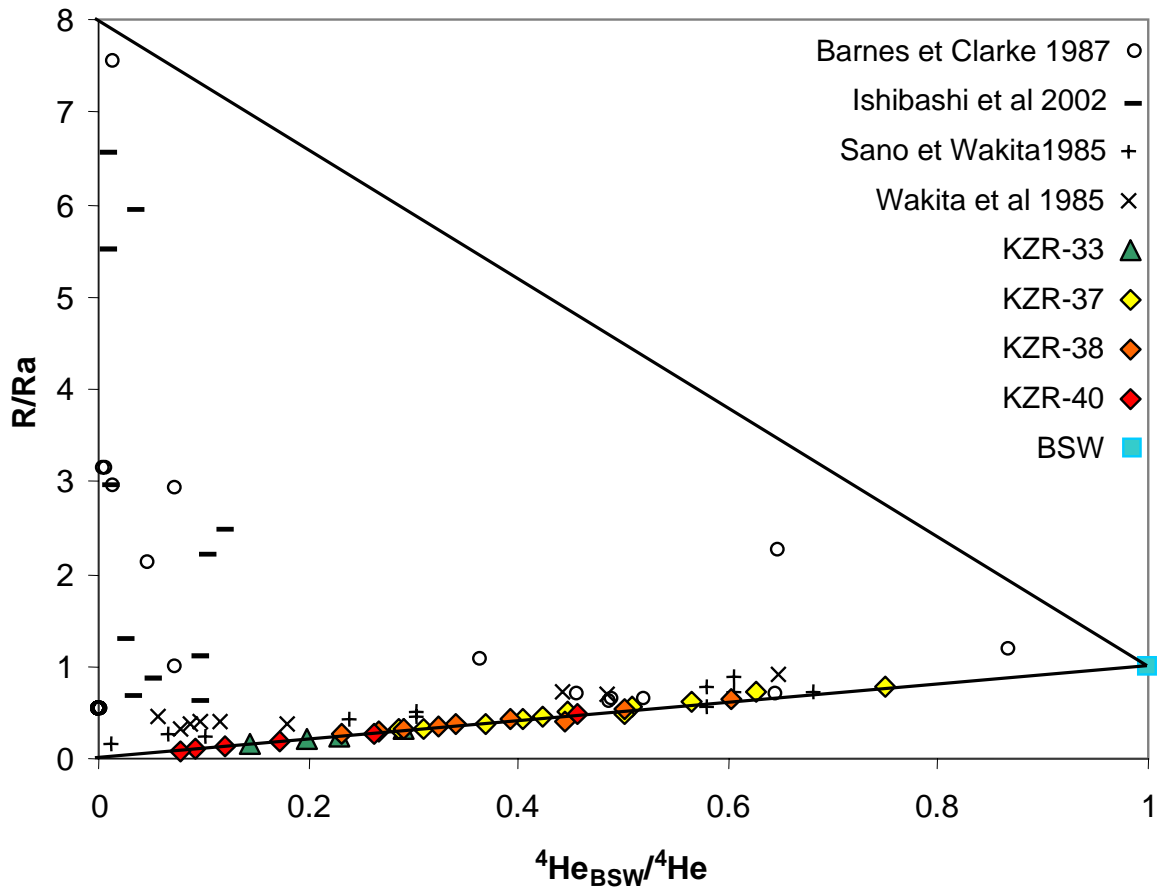


Fig. 7

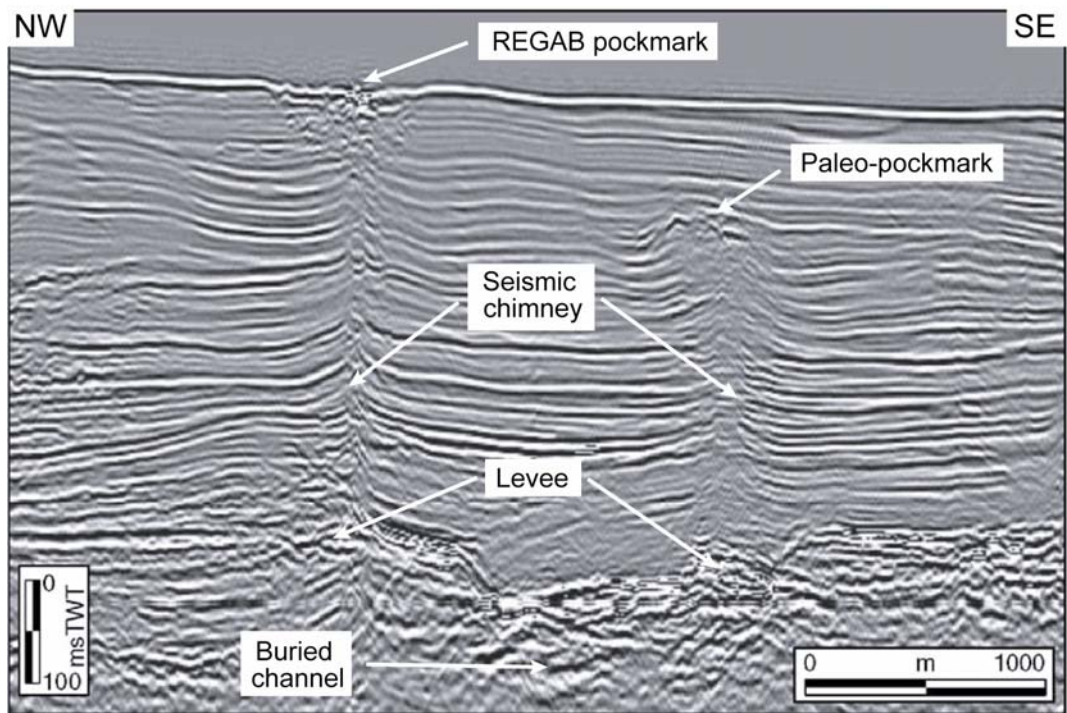


Fig. 8

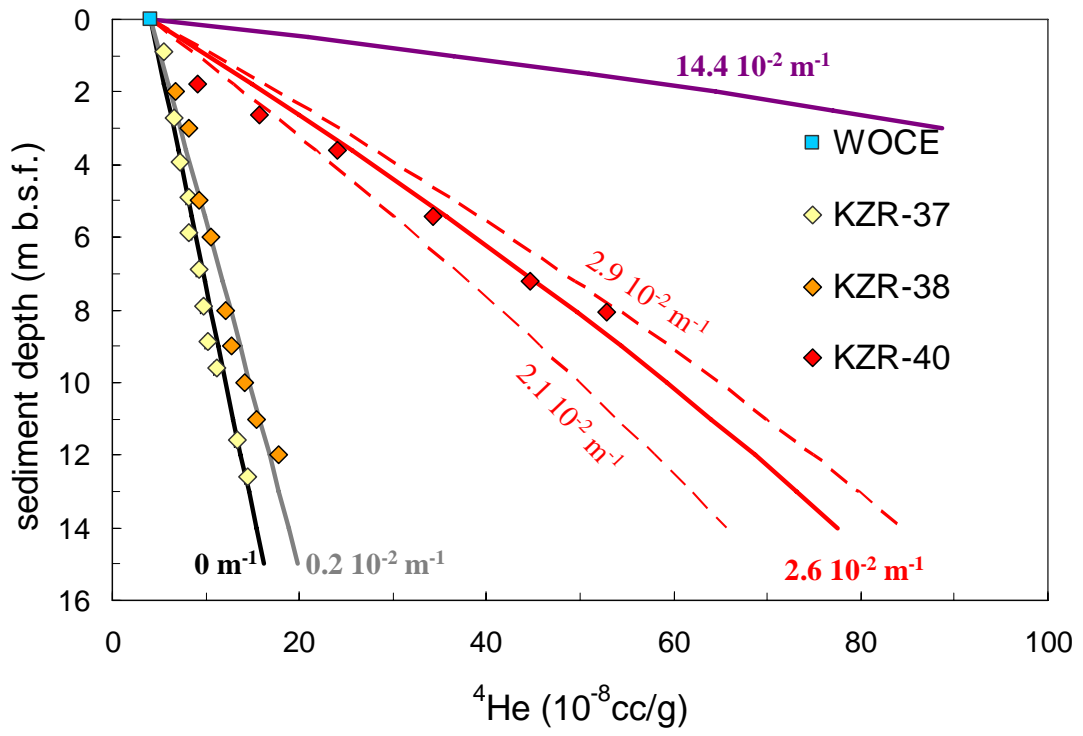


Fig. 9

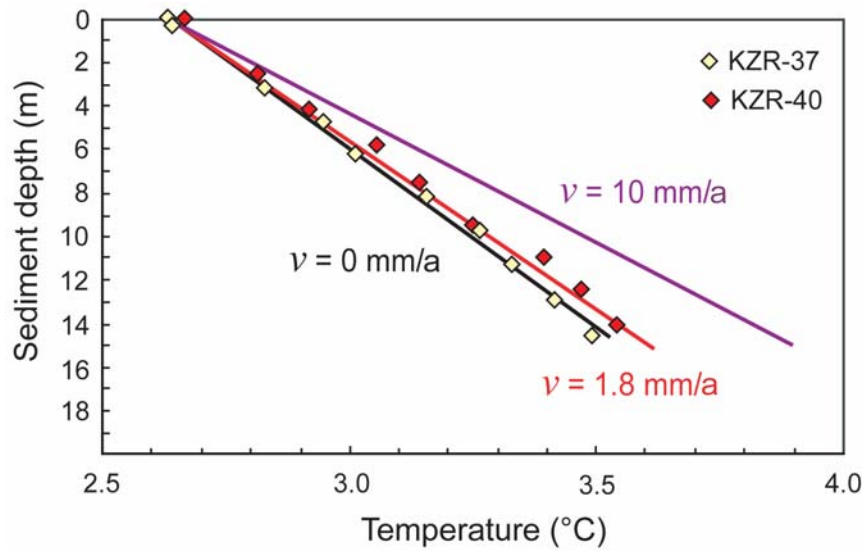


Fig. 10