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How precise are U-series coral ages?

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Abstract

U-series dating of fossil reef corals is a well established and widely applied technique in paleoclimate research. Many fossil corals, however, show evidence for post-depositional diagenetic alteration, and it is generally accepted that the accuracy of U-series coral ages is more limited due to coral diagenesis than analytical precision. In recent years, three models have been published that try to correct the effects of diagenesis and allow the calculation of model ages [Thompson W. G., Spiegelmann M. W., Goldstein S. L., and Speed R. C. (2003) An open-system model for U-series age determinations of fossil corals. *Earth and Planetary Science Letters* **210**, 365–381; Villemant B., and Feuillet N. (2003) Dating open systems by the ^{238}U–^{234}U–^{230}Th method: application to Quaternary reef terraces. *Earth and Planetary Science Letters* **210**, 105–118; Scholz D., Mangini A., and Felis T. (2004) U-series dating of diagenetically altered fossil reef corals. *Earth and Planetary Science Letters* **218**, 163–178].

Here, we assess the age variability of both conventional $^{230}Th/U$-dating and the three models by application to different sub-samples of individual coral specimens. The age variability, estimated as the 2σ-standard deviation on the individual ages, is compared with the errors quoted by the different methods. Our results show that the errors of conventional $^{230}Th/U$-dating as well as those of the method of Thompson et al. (2003) do not account for the true age variability. The age variability of both methods is in the range of the errors given by the models of Villemant and Feuillet (2003) and Scholz et al. (2004).

Furthermore, we show that the widely used reliability criteria are not sufficient to identify all diagenetically altered corals. In contrast, analysis of different sub-samples of one coral specimen allows (i) to estimate the real age variability, (ii) to test if the assumptions of the models are fulfilled, and (iii) to investigate the diagenetic processes in more detail. Thus, this method should generally be applied to obtain more reliable U-series coral ages and errors.

1. INTRODUCTION

The age/height relationship of fossil shallow water coral reefs represents a well established archive for past sea level fluctuations, and since the pioneering work of Mesolella et al. (1969) this reconstruction method has been widely applied (e.g., Fairbanks, 1989; Bard et al., 1996; Cutler et al., 2003). Because fossil corals can be dated by U-series methods (Edwards et al., 1987), such data provide an important independent test of the Milankovitch hypothesis of climate change (Milankovitch, 1941).

With respect to analytical uncertainty, very precise U-series ages can be obtained for fossil corals using mass spectrometric techniques (e.g., 2σ-errors of ± 1 thousand years (kyr) for 100 kyr-old samples, Edwards et al., 2003). However, most fossil corals (up to 90%, Thompson and Goldstein, 2005) display ($^{234}U/^{238}U$) activity ratios significantly higher than expected from closed system evolution of the ($^{234}U/^{238}U$) activity ratio measured on modern seawater ($(^{234}U/^{238}U)_{SW}$). Most values reported for $(^{234}U/^{238}U)_{SW}$ are around 1.146 (Chen et al., 1986; Cheng et al., 2000; Robinson et al., 2004a) and agree with the ($^{234}U/^{238}U$) activity ratio measured on modern corals. One study resulted in a slightly higher value (1.149 ± 0.002, (2σ − SD), Delanghe et al., 2002). All these values are calculated using the half-lives reported by Cheng et al. (2000). Although a recent study indicates shifts in

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$(^{234}U/^{238}U)_{SW}$ at times of major glacial-interglacial transitions involving large variations in sea levels (Esat and Yokoyama, 2006), several studies agree that $(^{234}U/^{238}U)_{SW}$ should not have varied more than 1% over glacial/interglacial cycles (Hamelin et al., 1991; Richter and Turekian, 1993; Robinson et al., 2004b). Because fractionation between different U isotopes does not occur during coral growth (Edwards et al., 2003), elevated $(^{234}U/^{238}U)$ activity ratios of fossil corals are clear evidence for coral open system behaviour. The isotopic anomalies are attributed to post-depositional diagenetic alteration, and the established opinion is that the accuracy of U-series coral ages is more limited due to diagenetic effects than analytical precision (Bard et al., 1992; Stirling et al., 1995).

Identification of diagenetically altered corals by mineralogical criteria is not possible because the U-series isotopic systems seem to be more sensitive to diagenetic change than any general geochemical parameter (Chen et al., 1991; Zhu et al., 1993). Thus, several criteria to identify reliable U-series coral ages have been developed (see e.g., Stirling et al., 1998). These are listed in Sections 2.1 and 3.1. An even more rigorous reliability test can be performed by combined $^{230}Th/U$- and $^{231}Pa/U$-dating (Edwards et al., 1997), and recent measurements revealed that six out of 14 corals with initial $(^{234}U/^{238}U)$, $(^{234}U/^{238}U)_{init.}$, within the range of the value measured on modern seawater had discordant $^{231}Pa/U$- and $^{230}Th/U$-ages (Gallup et al., 2002; Cutler et al., 2003) showing that this criterion alone is not sufficient to detect disturbed corals.

Various studies demonstrated the effects of post-depositional gain and loss of U and Th, respectively (e.g., Hamelin et al., 1991; Henderson et al., 1993), but for a long time none of these scenarios could be validated. Gallup et al. (1994) documented a rough trend in U-series coral data from Barbados, West Indies, on a $(^{234}U/^{238})$ vs. $(^{230}Th/^{238}U)$ plot. Based on this, they modelled the diagenetic processes assuming continuous addition of ^{234}U and ^{230}Th. This results in so-called addition lines which followed the trends in their data. Because the data scattered about the lines, Gallup et al. (1994) did not attempt to use their model to correct the U-series ages of their samples. A rather surprising aspect of this model is that ^{234}U and ^{230}Th are added at similar rates despite of their very different geochemical behaviour (i.e., U is soluble in natural waters while Th is not, Bourdon et al., 2003). Several authors have proposed mechanisms to solve this problem mainly involving particle reactive ^{234}Th, the recoil daughter of ^{238}U, which is likely to be hydrolysed and adsorbed onto solid surfaces before decaying to ^{234}U (Chen et al., 1991; Fruijtier et al., 2000).

In recent years, three models were developed to explain the isotopic anomalies in diagenetically altered corals (Thompson et al., 2003; Villemant and Feuillet, 2003; Scholz et al., 2004). In contrast to Gallup et al. (1994), the authors suggest that their models can be used to correct the U-series ages of coral samples with non-marine $(^{234}U/^{238}U)_{init.}$ and term these ages "open-system ages" (Thompson et al., 2003), "model ages" (Villemant and Feuillet, 2003), and "coral isochron ages" (Scholz et al., 2004), respectively. Meanwhile, these dating models have already been applied to derive paleoclimatic information (Felis et al., 2004; Potter et al., 2004; Thompson and Goldstein, 2005; Frank et al., 2006; Thompson and Goldstein, 2007).

To address questions like the timing and duration of past interglacials and interstadials (e.g., Stirling et al., 1998; Gallup et al., 2002) or the occurrence of suborbital sea level oscillations (Potter et al., 2004; Thompson and Goldstein, 2005), very accurate U-series ages of fossil corals are needed. As a consequence, the age errors must be reliably estimated. In most studies, the Th/U-age of a fossil coral is determined by analysis of its apparently best preserved part, but it is – of course – possible that one might have obtained a different age if another part of the coral had been used. Here, we determine the age variability of both conventional Th/U-ages and the different model ages by analysis of different sub-samples of a single coral specimen. This enables to check if the quoted errors account for the real geologic variation between the ages of different subsamples. Note that we do not question the applicability of the three models nor discuss their specific effects on coral age in this study because these questions were already addressed in detail in the original publications (Thompson et al., 2003; Villemant and Feuillet, 2003; Scholz et al., 2004) and elsewhere (Thompson and Goldstein, 2005; Frank et al., 2006; Scholz et al., 2006; Scholz and Mangini, 2007). Here, we focus on the variability of both conventional U-series coral ages and the ages calculated by the different models. The age variability of the different methods is calculated as the standard deviation on different sub-samples from an individual coral specimen. Our results show that the errors quoted for both the conventional and the open-system ages (Thompson et al., 2003) do not account for the real age variability.

2. SHORT DESCRIPTION OF THE MODELS

2.1. Conventional Th/U-dating

Conventional U-series dating of fossil corals is a well established technique for several decades. Thus, we do not go into the details here and only shortly summarise the general assumptions of the method. An extensive description is for instance given by Edwards et al. (2003).

The two general assumptions of Th/U-dating are that (i) initial $(^{230}Th/^{238}U) = 0$ and (ii) all changes in isotope ratios are the result of radioactive decay (i.e., no post-depositional chemical/diagenetic shifts in isotope ratios). While the first assumption is fulfilled for most modern surface corals (Edwards et al., 2003), the second assumption is violated for up to 90% of published coral data (Thompson and Goldstein, 2005). In most studies, fossil corals not satisfying the second assumption are identified by so-called "strict" reliability criteria: (i) $(^{234}U/^{238}U)_{init.}$ lying within a specific range around $(^{234}U/^{238}U)_{SW}$ (in the following referred to as the initial $(^{234}U/^{238}U)$ criterion), (ii) U concentrations comparable to modern corals of the same species, (iii) negligible ^{232}Th, and (iv) less than a specific amount of calcite (Stirling et al., 1998). In the following, these criteria are referred to as the standard screening criteria. We note that the exact values/

ranges used for the screening vary between individual studies. The uncertainty of conventional Th/U-ages is calculated by propagation of the analytical errors in (^{230}Th/^{238}U) and (^{234}U/^{238}U), respectively (see e.g., Ludwig (2003) for details).

2.2. The Thompson et al. (2003) model

This model assumes coupled addition (loss) of particle-reactive ^{230}Th and ^{234}Th (which rapidly decays to ^{234}U, $\tau_{1/2} = 24.1$ days) that are produced by decay of dissolved U and α-recoil mobilisation. These processes result in both increased (decreased) (^{234}U/^{238}U) and (^{230}Th/^{238}U) activity ratios, as observed in many fossil coral data. Based on this, Thompson et al. (2003) quantitatively modelled decay-dependent redistribution of ^{230}Th and ^{234}Th resulting in a modified age equation enabling the calculation of open-system coral ages for coral data plotting both above and below the seawater evolution curve (i.e., the curve describing the closed-system development of the activity ratios with (^{234}U/^{238}U)$_{init.}$ = (^{234}U/^{238}U)$_{SW}$).

Because the model only considers the redistribution process described above, corals that were affected by other diagenetic processes (e.g., U uptake/loss or presence of initial ^{230}Th) cannot be dated with the model and must be sorted out previously. This is done by application of standard screening criteria (see above) except for the initial (^{234}U/^{238}U) criterion. Open-system age errors are propagated from the analytical uncertainty in (^{230}Th/^{238}U) and (^{234}U/^{238}U) and random uncertainties in the model parameters (Thompson et al., 2003).

2.3. The Villemant and Feuillet (2003) model

Similarly as the Thompson et al. (2003) model, this approach assumes continuous selective redistribution of ^{234}U, ^{234}Th, and ^{230}Th controlled by α-recoil processes. Thus, it can be applied to coral data plotting both above and below the seawater evolution curve. In addition, possible initial ^{230}Th excess is taken into account and estimated using classical isochron diagrams (see e.g., Ludwig and Titterington, 1994). This implies that the model of Villemant and Feuillet (2003) can only be applied to a suite of coeval samples (i.e., at least two). This is a major difference to the Thompson et al. (2003) model and conventional Th/U-dating, which can be applied to a single coral measurement. Because the model does neither account for post-depositional U gain/loss nor for aragonite recrystallisation, samples that were affected by these processes must be sorted out using standard screening criteria (i.e., criteria (ii) and (iv), see above). The model age error is estimated using the standard deviations on the individual ages of the sample suite (Villemant and Feuillet, 2003).

2.4. The Scholz et al. (2004) model

This model assumes that different sub-samples of a single coral specimen gained different amounts of U with high (^{234}U/^{238}U), possibly followed by U loss proportional to U gain. This process results in a linear correlation between (^{230}Th/^{238}U) and (^{234}U/^{238}U), similarly as proposed by the models of Thompson et al. (2003) and Villemant and Feuillet (2003). In contrast to these models, however, the slope of the addition line (referred to as isochron) is not defined by the model but depends on the timing of U addition and loss, respectively, and therefore, on the specific diagenetic history of the analysed coral. Thus, the isochron is obtained from a linear fit of the data on a (^{234}U/^{238}U) vs. (^{230}Th/^{238}U) diagram. The coral isochron age is then calculated from the intersect of the isochron with the seawater evolution curve. The isochron model can also explain fossil coral data plotting below the seawater evolution curve if one assumes that the additional U has a lower (^{234}U/^{238}U) activity ratio than the coral.

Like the other models, coral isochron dating can only be applied if the underlying assumptions are fulfilled. For example, one can construct rather hypothetic scenarios with U loss being not proportional to U gain. In this case, the isochron age would not correspond to the true age of the coral (Scholz et al., 2004; Scholz and Mangini, 2007).

Coral isochron dating has been successfully applied to corals with high U content and elevated ^{232}Th but cannot be used to correct for aragonite recrystallization (Scholz et al., 2004). Thus, prior screening with criterion (iv) is required. The isochron age error is calculated by propagation of the isochron uncertainties (Scholz and Mangini, 2007).

3. METHODS AND MATERIAL

3.1. Methods

As explained in Section 2, there are several methods to determine the age of a fossil reef coral with U-series methods. In the case of conventional U-series dating, the apparently best preserved part of the coral is analysed. The age is then calculated from the measured activity ratios, and the age uncertainty is estimated by propagation of the *analytical uncertainties* (i.e., the errors derived from the counting statistics of the mass spectrometric analysis, the so-called *internal* or *within-run precision*). However, the internal errors often contribute only a part of the total measurement uncertainty and can, therefore, only be considered as a lower limit (Ludwig, 2003). The true precision of an analysis (i.e., the so-called *reproducibility* or *run-to-run precision*) can only be reliably established by replicate analysis of appropriate standard materials (Ludwig, 2003). For this reason, most laboratories do not use the analytical uncertainties if these are smaller than the evaluated reproducibility.

In the case of U-series dating of fossil reef corals, further problems arise from the effects of coral diagenesis. If the analysed coral part was altered by diagenetic effects, the determined age does, most likely, not correspond to the true age of the coral. The *accuracy* (i.e., the degree of conformity of the determined age to its true value) would then be much lower than suggested by both the analytical uncertainties and the reproducibility. To avoid this problem, it is common practice to identify diagenetically altered corals by the widely used reliability criteria, and if a sample fulfils all criteria, its age is believed to be accurate within the

assigned errors. It is, however, questionable if these criteria are sufficient to detect all altered corals. Here, we use measurements of different sub-samples of individual coral specimens to estimate the uncertainty of U-series coral ages, similar to the determination of the reproducibility by replicate analysis of standard materials. However, while all aliquots of such standard materials can reasonably be assumed to have the same isotopic composition, different sub-samples of one coral specimen can be affected by different degree of diagenesis or even different diagenetic processes. Thus, all sub-samples might have a significantly different isotopic composition within the limits of measurement reproducibility. For this reason, we refer to the uncertainty resulting from the analysis of different coral sub-samples as *age variability* and not age reproducibility. The age variability is calculated as the 2σ-standard deviation on the individual sub-sample ages. This is only possible for conventional ages and the ages calculated by the Thompson et al. (2003) model because only these methods allow to calculate an age for an individual analysis. The methods of Villemant and Feuillet (2003) and Scholz et al. (2004) can only be applied to a suite of coeval samples (i.e., sub-samples from an individual coral specimen or corals from the same stratigraphic unit).

The true ages of the individual sub-samples of a coral specimen should differ no more than a few hundred years. Thus, all calculated sub-sample ages should agree within the assigned errors. If this is not the case, this indicates that either the quoted errors were underestimated or the sub-samples cannot be dated with the respective model because they were affected by diagenetic processes that are not described by the model. As explained in Section 2, all models require prior screening for specific diagenetic effects that are not corrected by the model. Here, the following criteria are used:

(i) $(^{234}U/^{238}U)_{init.}$ must be in the range of $(^{234}U/^{238}U)_{SW}$ (i.e., between 1.140 and 1.152, Robinson et al., 2004a).
(ii) Total U concentration must be in the range of modern analogues. Here, we adopt the values used by Thompson et al. (2003) that are based on the results of Cross and Cross (1983). The value for *Porites* corals is based on measurements of modern specimens by Scholz et al. (2004). The U concentration ranges used for the different coral species are: *A. palmata*: 2.64–3.84 ppm; *M. annularis*: 2.12–3.32 ppm; *D. strigosa*: 2–3.2 ppm; *A. cervicornis*: 2.64–3.84 ppm; *Porites*: 2.1–3.8 ppm.
(iii) ^{232}Th content must be lower than 2 ppb.
(iv) Calcite content must be lower than 2%.

These widely used criteria are applied to all sub-samples (see electronic annex). In some cases, all sub-samples of a coral specimen pass the criteria. Then, all calculated sub-sample ages must be believed to be reliable. The mean sub-sample age then provides the best estimate of the coral age, and the age variability is calculated using all sub-sample ages. In other cases, only some sub-samples pass the criteria but others do not. Then, the age variability is calculated from those sub-samples that passed the criteria. Finally, there are several specimens, where all sub-samples fail the criteria. In this case, all ages cannot be considered to be reliable and the age variability cannot be assessed.

The age variability, which is considered as an estimate of the true uncertainty of the coral age, can be compared to the uncertainties quoted for both the conventional ages and the ages calculated by the Thompson et al. (2003) model. This comparison shows (i) if the quoted age errors do account for the total uncertainty and (ii) if the widely applied screening criteria are adequate to detect all altered sub-samples. In addition, the resulting age variability can be compared to the age errors estimated by the models of Villemant and Feuillet (2003) and Scholz et al. (2004).

All model calculations are performed with spreadsheets provided by the authors of the original publications using parameter values given there (Thompson et al., 2003; Villemant and Feuillet, 2003). Coral isochron ages and errors were calculated with the first order estimation method (Scholz and Mangini, 2007). Note that all models use slightly different values for $(^{234}U/^{238}U)_{SW}$. Thompson et al. (2003) use $(^{234}U/^{238}U)_{SW} = 1.145$, Scholz and Mangini (2007) $(^{234}U/^{238}U)_{SW} = 1.1466$, and Villemant and Feuillet (2003) $(^{234}U/^{238}U)_{SW} = 1.148$. These differences most likely result from the different values published for $(^{234}U/^{238}U)_{SW}$ (see Section 1). Because all model ages directly depend on the value used for $(^{234}U/^{238}U)_{SW}$, the absolute ages calculated by the different models should not be compared unless the uncertainty in $(^{234}U/^{238}U)_{SW}$ was considered (Scholz and Mangini, 2007). The error calculations of all models do not include this uncertainty. However, here we focus on the age variability. We only compare the ages each model calculates for different sub-samples but not the different model ages. For this reason, the usage of different values for $(^{234}U/^{238}U)_{SW}$ does not represent a problem.

3.2. Coral data

U-series measurements of different sub-samples of a single coral specimen are rare in the literature. Here, we use published U-series coral data from Barbados, West Indies (Gallup et al., 1994; Gallup et al., 2002; Thompson et al., 2003; Scholz et al., 2006), Aqaba, Jordan (Scholz et al., 2004), and the Huon Peninsula, Papua New Guinea (Esat et al., 1999), as well as new data from fossil corals collected on Barbados (Table EA1). U-series measurements of the new data were performed using thermal ionisation mass spectrometry (TIMS). Sample preparation and analysis are similar as described elsewhere (Scholz et al., 2006). All activity ratios shown in Table EA1 were calculated using the half-lives reported by Cheng et al. (2000), and older data (Gallup et al., 1994; Esat et al., 1999) were recalculated using these half-lives. The complete dataset consists of 38 coral specimens, and in most cases between two and four replicates from an individual specimen are available (Table EA1). For some specimens, however, a larger number of sub-samples (i.e., >8) is available (Table EA1). Most coral specimens grew during the Last Interglacial period (i.e., during Marine Isotope Stages (MIS) 5a, 5c, and 5e), but

the dataset also contains several older samples from MIS 6.5 and 7 (Table EA1).

XRD-measurements of the aragonite/calcite concentration are not available for all sub-samples listed in Table EA1 (Gallup et al., 1994; Esat et al., 1999; Scholz et al., 2006). Rejection of these samples would substantially decrease the size of the dataset. However, the sub-samples from the study of Scholz et al. (2006), which make up a large proportion of the dataset (Table EA1), were examined under ultraviolet light to identify parts that are mineralogically altered. Comparison with XRD-measurements revealed that the calcite fraction in these sub-samples appears whitish under ultraviolet light (Scholz et al., 2006). All sub-samples used here (Table EA1) showed no white portions indicating that their calcite content is negligible. Thus, we presume that these sub-samples fulfil criterion (iv) knowing that this represents a potential source of error.

As it was shown in previous studies, some of the coral data suffered a high degree of diagenetic alteration which is not only manifested in elevated $(^{234}U/^{238}U)_{init.}$ but also in increased U content (Scholz et al., 2004; Scholz et al., 2006). These data are, therefore, well qualified to (i) study the age variability these diagenetic effects produce in different sub-samples of the same specimen and (ii) to check if the widely applied screening criteria are sufficient to detect altered corals

4. RESULTS

4.1. Conventional ages

Fig. 1 shows a comparison between the age variability, calculated as the 2σ-standard deviation on the different sub-samples, and the mean of the quoted 2σ-errors of the conventional ages for the coral specimens listed in Table EA1. The average precision indicated by the quoted uncertainties is quantified by the mean value of the quoted errors. In the following we refer to this value shortly as the *mean error*. The histograms in Fig. 1 show the ratio between the age variability and the mean error in percent. Thus, all values larger than 100% indicate that the age variability is larger than the quoted errors suggest, while values lower than 100% indicate that the age variability is smaller. In Fig. 1a all 38 coral specimens from Table EA1 are included. Only seven specimens have a ratio lower than 100%, and the mean ratio is 622% suggesting that the quoted errors do not account for the true variability in the ages. However, large values may be explained by diagenetic alteration because several sub-samples do not fulfil criteria (i)–(iv) (Table EA1). Fig. 1b shows the ratios after screening with criteria (ii)–(iv), which should eliminate all sub-samples that suffered U gain/loss (ii), were affected by detrital Th (iii) and/or were recrystallized (iv). Because in some cases all sub-samples of a coral specimen do not meet the criteria, the total size of the dataset is reduced to 32 (see Section 3.1). For example, coral specimen BB02-5-4 which shows clear signs of both U loss and recrystallisation (Table EA1) is sorted out by this screening. However, the mean ratio is still 528% (Fig. 1b). Fig. 1c shows the ratios after

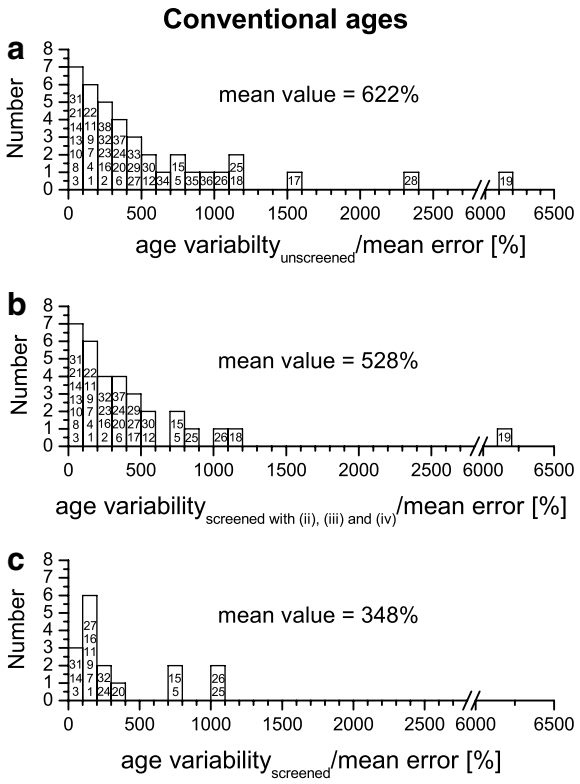

Fig. 1. Histograms showing the ratio between the age variability and the mean error of the conventional ages of the different sub-samples (Table EA1). The numbers within the bins correspond to the respective coral specimens (Table EA1). Panel (a) shows all coral specimens (age $\text{variability}_{unscreened}$/mean error), panel (b) shows all specimens that passed the screening with criteria (ii), (iii), and (iv) (age $\text{variability}_{screened\ with(ii),(iii)\ and\ (iv)}$/mean error), and panel (c) shows the screened dataset (age $\text{variability}_{screened}$/mean error).

further screening with criterion (i) (i.e., $(^{234}U/^{238}U)_{init.}$). This reduces the total sample size to 16 specimens. Because these specimens passed all criteria, their ages must be considered as reliable. However, only three of these 16 specimens have a ratio lower than 100% indicating that the quoted error accounts for the age variability. In contrast, seven specimens have a ratio larger than 200% suggesting that the true age variability is more than twice as large as the quoted error. The mean ratio after screening with all criteria is still 348%. This shows that either the widely used criteria are not sufficient to eliminate all samples affected by diagenesis or the quoted errors were substantially underestimated.

4.2. Open-system ages (Thompson et al., 2003)

Because the Thompson et al. (2003) model can be applied to a single coral measurement, the open-system age variability can be estimated as the 2σ-standard deviation on the sub-sample open-system ages, as described in Section 3.1. The results are presented in Fig. 2. The Thompson et al. (2003) model does not correct for post-depositional U uptake/loss, initial presence of ^{230}Th nor aragonite

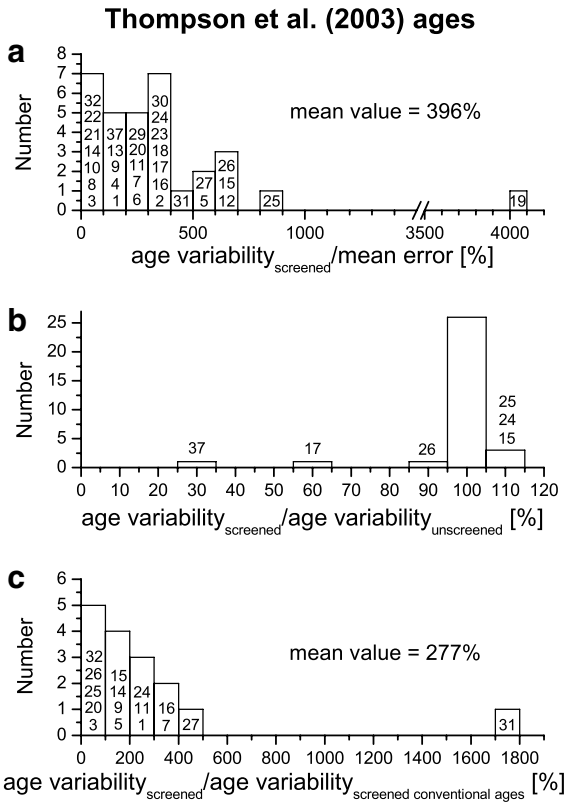

Fig. 2. The histogram in Panel (a) shows the ratio between the age variability and the mean error of the open-system ages (Thompson et al., 2003) in percent. To eliminate corals that were altered by diagenetic processes not included in the model, the data were screened with criteria (ii), (iii) and (iv). Panel (b) shows the ratio between the variability of the screened and the unscreened open-system ages in percent. The histogram in panel (c) shows the ratio between the age variability of the screened open-system ages and the screened conventional ages in percent. The numbers within the bins correspond to the respective coral specimens (Table EA1).

recrystallisation. Thus, sub-samples that possibly were affected by these processes must be sorted out with criteria (ii)–(iv). Fig. 2a presents the comparison between the open-system age variability and the mean quoted open-system age errors. The histogram shows the ratio between the age variability and the mean error in percent. Screening with criteria (ii)–(iv) eliminates six of the 38 coral specimens (Table EA1) because none of the sub-samples fulfils all criteria (see above). Only seven of the remaining 32 coral specimens have a ratio lower than 100% (Fig. 2a), but 20 corals have a ratio larger than 200%. The mean value is 396%. This indicates, similarly as for the conventional ages, that either (i) the widely used criteria are not adequate to identify all altered corals, or (ii) the quoted uncertainty is substantially underestimated, or (iii) the Thompson et al. (2003) model is not applicable to these coral data.

The histogram in Fig. 2b shows the ratios between the age variability of the screened and the unscreened open-system ages in percent. Values around 100% indicate that the screening does not change the age variability, which is the case if all sub-samples of a coral specimen pass all criteria. Smaller ratios indicate an error improvement due to the screening, while larger values suggest that the error is enlarged. The large peak at 100% (26 counts, Fig. 2b) shows that the screening does not have a major effect on the observed open-system age variability. In most cases, either all sub-samples pass the screening or all sub-samples fail so that the whole specimen is eliminated. The age variability is only for six specimens affected by the screening: Three specimens show a lower ratio after the screening, and three show a slightly larger (Fig. 2b).

Fig. 2c shows the comparison between the age variability of the screened open-system ages (Thompson et al., 2003) and the screened conventional ages. The histogram contains only 16 specimens because the other corals are eliminated by the screening (compare Fig. 1c). In the majority of cases the age variability of the open-system ages (Thompson et al., 2003) is larger than that of the conventional ages. Only five specimens show a ratio lower than 100% indicating that the variability of the open-system age (Thompson et al., 2003) is smaller. For the other eleven specimens the variability of the open-system ages (Thompson et al., 2003) is larger than that of the conventional ages. The mean ratio is 277% suggesting that, in average, the variability of the screened open-system ages is about three times larger than that of the screened conventional ages.

4.3. Model ages (Villemant and Feuillet, 2003)

In contrast to conventional U-series coral dating and the Thompson et al. (2003) model, the Villemant and Feuillet (2003) model can only be applied to a suite of samples because classical isochron methods are involved. However, the model calculates an individual age for each sub-sample so that the age variability (i.e., the standard deviation) can be calculated. Indeed, Villemant and Feuillet (2003) use the standard deviation on the individual ages to estimate the error of their model ages.

The Villemant and Feuillet (2003) model can be used to correct for initial ($^{234}U/^{238}U$) activity ratios that cannot be explained by closed-system decay and for initial presence of ^{230}Th (as indicated by elevated ^{232}Th content). It does neither account for post-depositional U uptake/loss nor for aragonite recrystallisation. Thus, the coral data (Table EA1) were screened with criteria (ii) and (iv) before the model was applied which results in rejection of three coral specimens. If only one sub-sample passes the screening, it is not possible to calculate the standard deviation. This is the case for two coral specimens.

Fig. 3 shows the comparison of the quoted 2σ-errors of the Villemant and Feuillet (2003) model with the age variability of the conventional and the open-system ages (Thompson et al., 2003). It is evident that the variability of the Villemant and Feuillet (2003) ages is generally larger than the variability of the conventional ages (Fig. 3a). Only two specimens have a ratio lower than 100%. In contrast, eight specimens have a ratio larger than 200% indicating a substantially larger variability of the Villemant and Feuillet (2003) ages. The mean ratio is 376%.

Because the model of Villemant and Feuillet (2003) assumes similar redistribution mechanisms as the Thompson

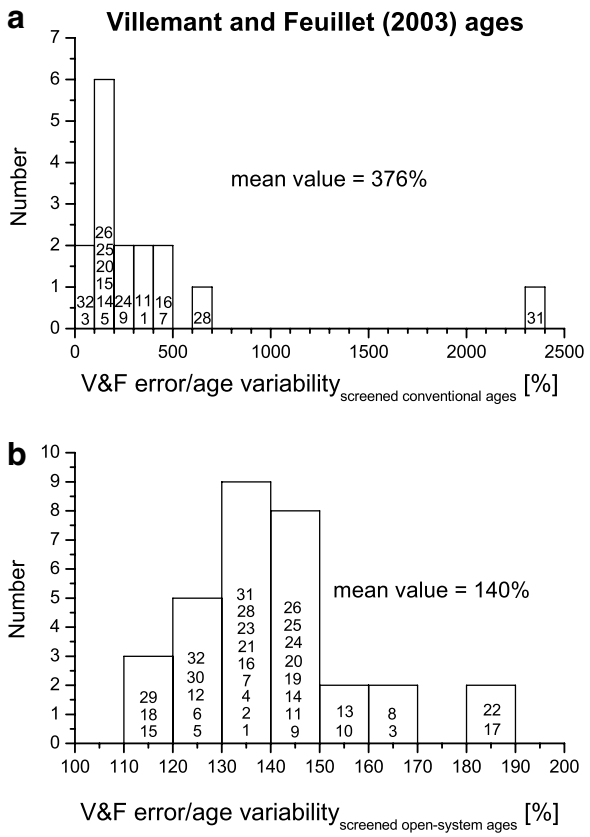

Fig. 3. Histograms showing the ratios between the errors of the Villemant and Feuillet (2003) model and the age variability of the conventional and open-system ages (Thompson et al., 2003), respectively. The numbers within the bins correspond to the respective coral specimens (Table EA1). Panel (a) shows the comparison with the conventional ages and panel (b) that with the Thompson et al. (2003) ages.

et al. (2003) model (see Section 2), the ages calculated by the two models differ only slightly. Thus, it is not surprising that the errors of the Villemant and Feuillet (2003) ages (that are calculated as the standard deviation on the individual sub-sample ages) and the age variability of the open-system ages (Thompson et al., 2003) are approximately of the same magnitude (Fig. 3b). Interestingly, the variability of the Villemant and Feuillet (2003) ages is generally larger (Fig. 3b), which is probably due to minor differences between the two models or due to the different values that were used for $(^{234}U/^{238}U)_{SW}$ (see Section 3.1). However, the ratios are all between 110% and 190%, and the mean ratio is 140% indicating that the differences are slight.

Note that these results imply that the errors calculated by the Villemant and Feuillet (2003) model are substantially larger than the errors quoted for both the conventional ages and the open-system ages that are based on the analytical uncertainties only (Figs. 1 and 2). However, considering the age variability of both the conventional and the open-system ages, the errors quoted for the Villemant and Feuillet (2003) ages seem to be more realistic.

4.4. Coral isochron ages (Scholz et al., 2004)

The uncertainty of coral isochron ages is estimated by propagation of the isochron errors (i.e., the uncertainty of the linear fit of the coral data on a $(^{234}U/^{238}U)$–$(^{230}Th/^{238}U)$-diagram, Scholz and Mangini, 2007). The errors shown in Fig. 4 were calculated with the first order estimation method (Scholz and Mangini, 2007). In contrast to the three other models, it is not possible to calculate an isochron age for a single coral sub-sample but only for a suite of sub-samples. Thus, it is not possible to calculate the age variability (i.e., the standard deviation) for the coral isochron ages, as described for the other methods. However, if the coral data do not show a linear correlation on a $(^{234}U/^{238}U)$–$(^{230}Th/^{238}U)$-diagram, which is a clear indication for a deviation from the model assumptions, this is considered by the coral isochron age error calculation, and the error is enlarged appropriately (Scholz and Mangini, 2007).

Coral isochron dating can be used to correct post-depositional U uptake/loss resulting in $(^{234}U/^{238}U)_{init.}$ activity ratios different from $(^{234}U/^{238}U)_{SW}$ and was also successfully applied to corals with elevated ^{232}Th (Scholz et al., 2004). Thus, it is only necessary to screen for aragonite recrystallisation with criterion (iv), which eliminates two coral specimens from Table EA1. However, because at least three sub-samples are necessary to calculate an isochron age error (Scholz and Mangini, 2007), isochron dating cannot be applied to specimens with less than three sub-samples (Table EA1). This eliminates eleven other specimens. As noted above, the coral isochron age errors become large if the data points scatter about the isochron and/or the data point distances are small (Scholz and Mangini, 2007). In this case, the isochron dating assumptions are assumed to be not fulfilled, and the method should not be applied. Thus, only isochron ages with a relative error lower than 15% are used here. This eliminates twelve other specimens. Finally, ten

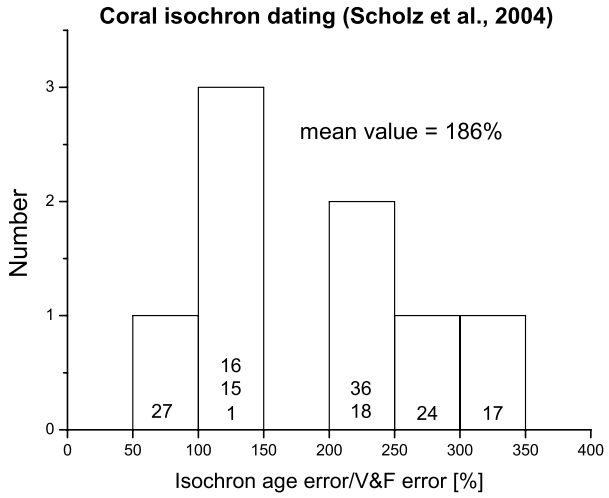

Fig. 4. Histogram showing the ratios between the error of the coral isochron ages (Scholz et al., 2004) and the Villemant and Feuillet (2003) ages in percent. The numbers within the bins correspond to the respective coral specimens (Table EA1).

specimens remain, which can be dated with coral isochron dating.

Fig. 4 shows the comparison between the coral isochron age errors and the uncertainties of the Villemant and Feuillet (2003) ages. Only one of these specimens has a ratio lower than 100%. In contrast, four specimens have a ratio larger than 200% indicating that the isochron age error is more than twice as high as the Villemant and Feuillet (2003) age error. The mean ratio is 186%. Note that only isochron ages with an error smaller than 15% are included in Fig. 4 (see above) and many others have even larger errors (Table EA1). This of course implies that the coral isochron age errors are also substantially larger than the uncertainties quoted for the conventional and the open-system ages (Thompson et al., 2003), which are much smaller than the Villemant and Feuillet (2003) age errors (see Section 4.3. and Fig. 3).

5. DISCUSSION

Shortly summarized, the results of the previous section show that the uncertainties quoted for both the coral isochron ages (Scholz et al., 2004) and the model ages (Villemant and Feuillet, 2003) are substantially larger than those given for the conventional and the open-system ages (Thompson et al., 2003). However, the real age variability of the conventional and open-system ages (Thompson et al., 2003), estimated as the 2σ-standard deviation of different sub-samples ages of a single coral specimen, seems to be in the range of the uncertainty of the Villemant and Feuillet (2003) and the coral isochron ages (Scholz et al., 2004). Because both the Villemant and Feuillet (2003) and the Scholz et al. (2004) age errors are calculated using the scatter of the individual sub-samples, this is not surprising. It is much more important, however, that the age variability of both the conventional and the Thompson et al. (2003) ages is substantially larger (i.e., between 3 and 4 times) than the quoted errors suggest (Figs. 1c and 2a), even for coral specimens, which pass all screening criteria.

The discrepancy between the quoted errors and the true age variability may have two reasons: Firstly, the measurement uncertainties might have been underestimated, which may be possible considering the small errors resulting from mass spectrometric analyses. This, however, should not be the case because the uncertainty of mass spectrometric measurements is normally not estimated from the in-run counting statistics but from the reproducibility of replicate measurements of an appropriate standard material (Ludwig, 2003; see also Section 3.1). The second possibility is that the coral samples with large ratios between age variability and mean quoted error (i.e., larger than 200%, see Figs. 1 and 2 and Table EA1) were affected by diagenetic processes that are not accounted for by the respective model. This would mean that these corals should not be dated with the models because the calculated ages had no significance. Such sub-samples, however, are assumed to be sorted out previously by the screening. Maybe the widely used criteria are not strict enough, but it is also possible that such criteria are generally not adequate to identify all sub-samples that were affected by diagenetic processes not considered by the models. Because other studies used exactly the same criteria (Thompson et al., 2003; Thompson and Goldstein, 2005) and their failure would, thus, also pertain to the results of these studies, it is important to investigate this question in more detail.

To test, if the, in part, very large ratios between the age variability and the errors quoted for the conventional (Fig. 1c) and the Thompson et al. (2003) ages (Fig. 2a) are produced by diagenetically altered sub-samples that erroneously passed the screening, we redefine the criteria. The $(^{234}U/^{238}U)_{init.}$ range (see point (i) in Section 3) is modified to $1.143 < (^{234}U/^{238}U)_{init.} < 1.149$. Similarly, the range of U content for the individual coral species (see point (ii) in Section 3) is halved. This results for example in a 'new' range of 2.94–3.54 ppm for *Acropora palmata* corals. Finally, only sub-samples with a ^{232}Th content less than 1 ppb and less than 1% calcite are assumed to be reliable. Application of these (stricter) criteria of course results in rejection of more sub-samples. After application of the stricter criteria, the ratio between the age variability and the mean errors of the conventional ages can just be calculated for nine coral specimens. These ratios are presented in the histogram in Fig. 5a. Comparison with Fig. 1c reveals that the screening with the stricter criteria results in the rejection of the coral specimens with the largest ratios (i.e., BB02-5-1 (No. 25), BB02-5-2 (No. 26) and GQ-3 (No. 5)). Nevertheless, still only one coral specimen has a ratio lower than 100% suggesting that the quoted error is adequate to account for the real age variability, while four specimens have ratios larger than 200% indicating that the age variability is more than twice as high as the quoted error (Fig. 5a). The mean ratio is 259%. This shows that the application of stricter criteria slightly reduces the discrepancy between the age variability and the quoted errors, but is not adequate to reject all samples that exhibit more variability than suggested by the analytical errors.

The results for the open-system-ages (Thompson et al., 2003) are similar. The ratio between the age variability and the mean error can be calculated for 23 coral specimens after the modified screening. These ratios are presented in the histogram in Fig. 5b. As for the conventional ages, the specimens with the largest ratios (compare Fig. 2a) are rejected by the stricter criteria (i.e., specimens BB02-5-1 (No. 25), BB02-5-2 (No. 26) and ADU-2 (No. 19)). In contrast, the ratio of specimen BB02-5-3 (No. 27) is enlarged from 264 to 331%. This reveals that the application of stricter criteria does not always result in a smaller ratio between age variability and mean error. Five coral specimens have a ratio lower than 100%, while 15 have a ratio larger than 200% (Fig. 5b). The mean ratio is 272% showing that the application of stricter criteria reduces the discrepancy between the age variability and the quoted errors to some extent. However, in the majority of cases, the real age variability is still much larger than the quoted errors, similarly as for the conventional ages.

Fig. 5c shows the comparison between the age variability of the open-system (Thompson et al., 2003) and the conventional ages after the stricter screening. The histogram contains only nine specimens because all other corals are eliminated by the screening (compare Fig. 5a). In the

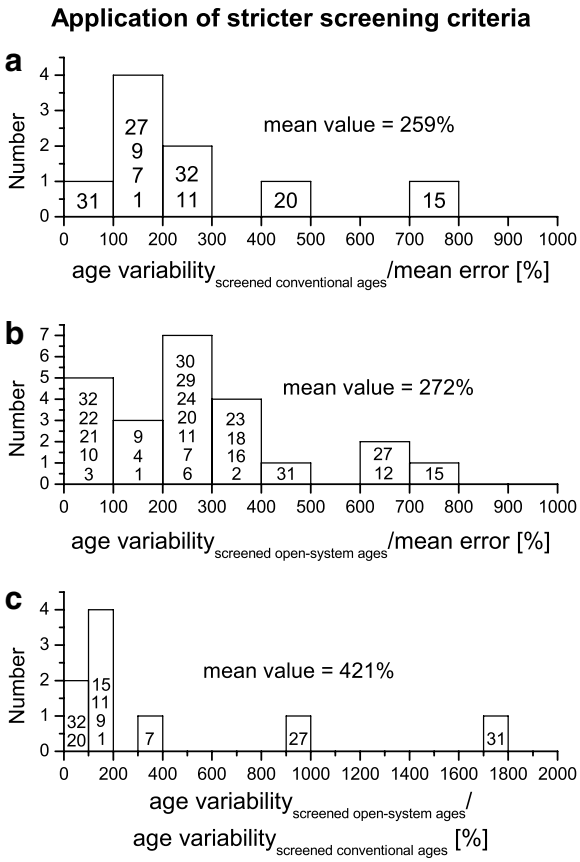

Fig. 5. (a) Histogram showing the ratio between the age variability and the mean error of the conventional ages in percent after screening with the stricter criteria (see main text for details). (b) Histogram showing the ratio between the age variability and the mean error of the open-system-ages (Thompson et al., 2003) in percent after screening with the stricter criteria. (c) Histogram showing the ratio between the age variability of the open-system ages (Thompson et al., 2003) and the conventional ages in percent. The numbers within the bins correspond to the respective coral specimens (Table EA1).

majority of cases, the variability of the open-system ages (Thompson et al., 2003) is larger than that of the conventional ages. Only two specimens have a ratio lower than 100%. The mean ratio is 421% suggesting that, in average, the variability of the open-system ages (Thompson et al., 2003) is about four times larger than that of the conventional ages.

The discussion above shows that the application of stricter screening criteria helps to reduce the ratio between the age variability and the quoted errors to some extent for both the conventional and the open-system ages (Thompson et al., 2003). However, many coral specimens still have ratios larger than 200% (~45% of the conventional ages, Fig. 5a, and ~65% of the open-system ages, Fig. 5b). If we presume that the quoted errors were carefully assessed (i.e., the reproducibility was determined using replicate measurements of standard materials) and, thus, exclude that they were substantially underestimated, this can only be explained by diagenetic processes that are not detected by the screening. In the case of the conventional ages, this simply means that the coral did not behave as a closed system. In the case of the open-system ages (Thompson et al., 2003), it means that the coral was affected by other diagenetic processes than assumed by the model. It is questionable if the widely used screening criteria are the best method to identify sub-samples which suffered diagenetic alteration because the application of stricter criteria does not only result in rejection of the specimens with the largest ratios but also of specimens with ratios lower than 100% (compare on this Figs. 5a and 1c and also Figs. 5b and 2a). That screening criteria are not sufficient to identify all altered samples was already confirmed by coral samples with discordant ^{231}Pa/U- and ^{230}Th/U-ages which, however, fulfilled the reliability criteria applied by the authors (Gallup et al., 2002; Cutler et al., 2003).

At present, there are two other possibilities than screening criteria to detect diagenetically altered corals : (i) combined ^{230}Th/U- and ^{231}Pa/U-dating (Edwards et al., 1997; Gallup et al., 2002; Cutler et al., 2003; Chiu et al., 2006) and (ii) analysis of several sub-samples of an individual coral specimen. The second method has, apart from detecting diagenetically altered samples, three further advantages: (i) It may be possible to obtain additional information on the diagenetic processes that affected the coral, for example post-depositional U-uptake/loss (Scholz et al., 2004; Scholz et al., 2006). (ii) It is possible to check if the assumptions of the different models are fulfilled. For example, one can compare possible data trends with those predicted by the Thompson et al. (2003) model on a (^{234}U/^{238}U)–(^{230}Th/^{238}U)-diagram (Scholz et al., 2006). (iii) The age variability of both the conventional and the open-system ages can be estimated as described in this study. If the age variability is substantially larger than the quoted uncertainty, the analytical error can be enlarged appropriately to avoid underestimation of the true error. These three points highlight the advantages of analysing several sub-samples of an individual coral specimen.

Of course, one could ask if it is justified to use all coral specimens listed in Table EA1 for this study because it was already shown that some of them were altered by different diagenetic processes than assumed by the models. For example, if the sub-samples do not show a correlation between (^{234}U/^{238}U) and (^{230}Th/^{238}U) or the trend line has a significantly different slope than presumed by the models of Thompson et al. (2003) and Villemant and Feuillet (2003), one could argue that these models should not be applied to the data. This is reasonable because it indicates that the model assumptions are not fulfilled, and the calculated model ages would be without significance. It is, however, only possible to test if the activity ratios are correlated and if the slope agrees with that proposed by the models if at least three different sub-samples of a coral specimen are analysed. The Thompson et al. (2003) model and conventional U-series dating, however, are in most cases applied to a single measurement of a coral. The problems arising from this practice are exemplarily illustrated with corals AEC 1 and AFM 6 (Thompson et al., 2003; Table EA1). Figs. 6 and 7 show (^{234}U/^{238}U) vs. (^{230}Th/^{238}U) diagrams for both corals. In case of coral AEC 1 (Fig. 6), all

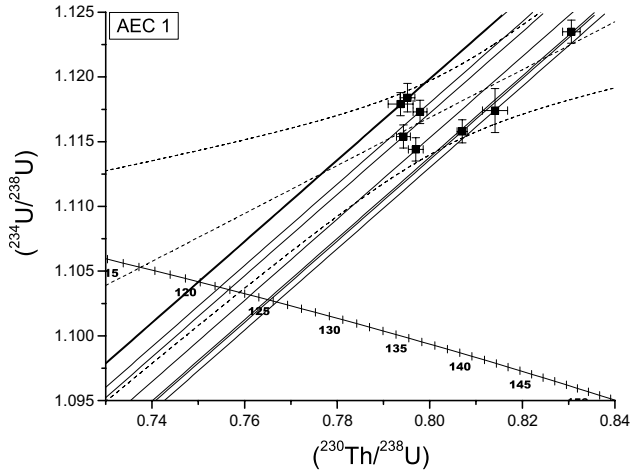

Fig. 6. ($^{234}U/^{238}U$) vs. ($^{230}Th/^{238}U$) diagram for coral AEC 1. The straight curve is the seawater evolution curve and the ticks and numbers indicate the corresponding U-series age in kyr. The dashed lines are the isochron obtained from a linear fit through the data-points and the corresponding confidence bands. The methods that were used for the calculation of the isochron and the confidence bands are described in detail in Scholz and Mangini (2007). The isochron age (Scholz et al., 2004; Scholz and Mangini, 2007) can be obtained from the intersect of the isochron with the seawater evolution curve. The parallel solid straight lines intersecting each data-point are the trend lines predicted by the Thompson et al. (2003) model. The open-system ages (Thompson et al., 2003) can be obtained from the intersects of these trend lines with the seawater evolution curve.

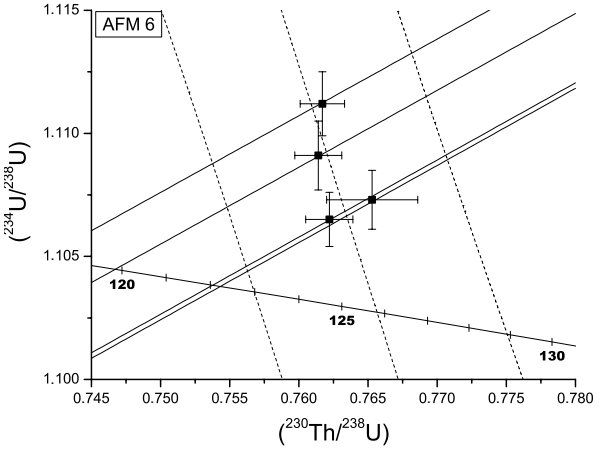

Fig. 7. ($^{234}U/^{238}U$) vs. ($^{230}Th/^{238}U$) diagram for coral AFM 6. The notation is analogous to Fig. 6.

sub-samples have elevated $(^{234}U/^{238}U)_{init.}$, and their conventional ages are, therefore, assumed to be not reliable. The slope of the fit through the data points (dashed line) is in the range of the slope predicted by the Thompson et al. (2003) model (Fig. 6), even if four of the Thompson et al. (2003) trend lines plot outside the 2σ-confidence bands. Because of the rather large error of the isochron age (117.1 ± 11.4 kyr, Table EA1), it is in agreement with the single ages calculated by the Thompson et al. (2003) model, which range from 120.1 ± 1.2 to 125.6 ± 1.8 kyr (Fig. 6, Table EA1). However, it is evident that not all sub-samples plot on one Thompson et al. (2003) trend line (Fig. 6). Instead, each sub-sample seems to define its own trend line (Fig. 6). In consequence, each sub-sample yields its own open-system age (Thompson et al., 2003), and, due to the small quoted errors, the age differences between the individual sub-samples are significant. This shows that the assumptions of the Thompson et al. (2003) model cannot be fulfilled for coral AEC 1. But which of the open-system ages should be used? Is the true coral age in the range of 120 kyr or rather in the range of 125 kyr? Because there are several analyses available, it is possible to use the mean of the open-system ages as an estimate of the true open-system age, and as described in Section 3, the open-system age variability should represent a reasonable estimate of the age uncertainty. However, if only a single sub-sample had been analysed, the open-system age and the quoted errors would have been believed to be reliable. In other words, the resulting open-system age would be between 120 and 125 kyr, depending on the coral part that was analysed. In the case of coral AFM 6 (Fig. 7), the slope of the linear fit through the data-points (dashed line) is negative and not in agreement with the slope predicted by the Thompson et al. (2003) model showing that the model assumptions are not fulfilled. Again, each sub-sample plots on its own trendline, and the corresponding open-system ages (Thompson et al., 2003) are between 117.6 ± 1.1 and 121.5 ± 1.2 kyr (Table EA1). In contrast, the conventional ages of sub-samples AFM 6-0 and –1, which pass all criteria, are 124.7 ± 1.0 and 123.9 ± 0.6 kyr (Table EA1) suggesting an older coral age. In summary, these examples highlight the necessity to analyse several sub-samples of one coral specimen and, at the same time, reveal the problems of the application of the Thompson et al. (2003) model to single coral measurements from the literature (Thompson and Goldstein, 2005; Thompson and Goldstein, 2007). The results of this study indicate that the published open-system ages (Thompson and Goldstein, 2005; Thompson and Goldstein, 2007) are associated with much larger uncertainty.

As visible in Figs. 2c and 5c, the age variability of the open-system ages is between three and four times larger than the variability of the conventional ages. This is contrary to other studies (Thompson et al., 2003; Thompson and Goldstein, 2005; Frank et al., 2006), which suggest that the application of the Thompson et al. (2003) model reduces the age variability between different corals from the same stratigraphic unit. This apparent discrepancy may arise from the fact that Figs. 2c and 5c contain only sub-samples that fulfil all screening criteria, which implies that these samples are not strongly altered. In other words, sub-samples, for which a model application would have a large effect, are not contained in Figs. 2c and 5c. However, as explained before, model application to corals from the same stratigraphic unit is a less rigorous test than application to different sub-samples from an individual coral specimen. While the latter are definitely of the same age, age variations between different corals from one reef may reflect real age differences or arise from a failure of the model. In Fig. 8, the age variability of both the conventional and the open-system ages (Thompson et al., 2003) is plotted against the standard deviation of $\delta^{234}U(\sigma_{\delta^{234}U})$, which is a measure for the variability of ($^{234}U/^{238}U$) in different sub-samples. The upper panel of Fig. 8, which shows the conventional ages, displays a rough trend suggesting increasing age variability with larger $\sigma_{\delta^{234}U}$. Assuming that $\sigma_{\delta^{234}U}$ is a measure for the variability of the degree of diagenetic alteration in different sub-samples, it is reasonable that the age variability in these sub-samples (which is assumed to be produced by coral diagenesis) is also large. If the Thompson et al. (2003) model corrected these diagenetic effects, there should be no trend visible between the age variability of the open-system ages (Thompson et al., 2003) and $\sigma_{\delta^{234}U}$. However, as evident from the lower panel of Fig. 8, there is a similar trend in the age variability of the open-system ages clearly showing that the Thompson et al. (2003) model does not correct the diagenetic effects, which affected these corals. Especially for those specimens with the largest age variability (corals No. 19, 25, 26, 27 and 29, Fig. 8), the model application seems to have no effect. Because Fig. 8 contains only sub-samples which passed criteria (ii), (iii) and (iv), other diagenetic processes like U uptake/loss should play a minor role. This analysis, which is based on 32 coral specimens (Table EA1), confirms that the application of the Thompson et al. (2003) model in many cases increases the age variability instead of correcting the diagenetic effects (Figs. 2c and 5c). At this stage, it remains an open question why the Thompson et al. (2003) model enlarges the age variability of different sub-samples from one coral specimen but reduces the age variability of different corals from one stratigraphic unit. A possible explanation may be as follows: For corals with elevated ($^{234}U/^{238}U$), the Thompson et al. (2003) model always results in an age reduction. Because diagenetic alteration normally produces both elevated ($^{234}U/^{238}U$) and age, the Thompson et al. (2003) model shifts the ages of the altered corals towards the age range of the unaltered corals. This of course involves a reduction of the age variability within the reef. However, this works for all corals, which display a trend between ($^{234}U/^{238}U$) and age, and not only for corals, which have been altered

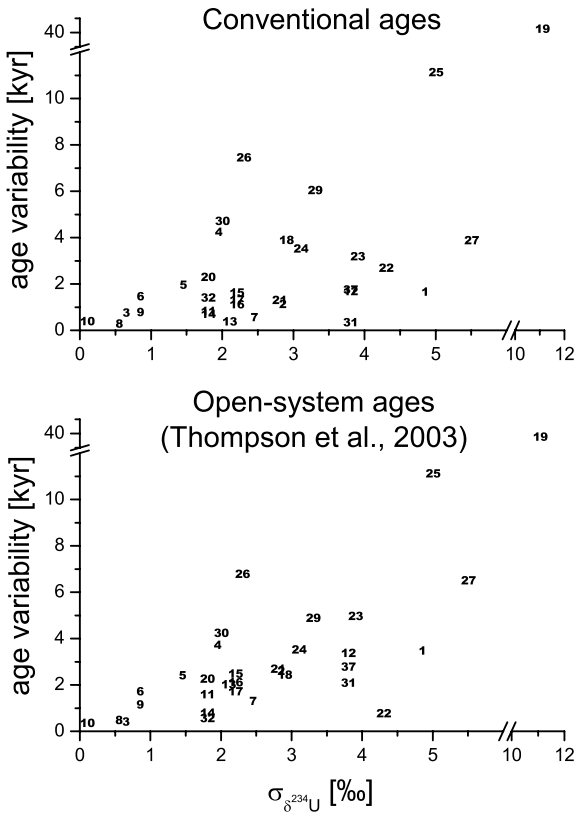

Fig. 8. Age variability vs. standard deviation of $\delta^{234}U$ diagrams for both the conventional (upper panel) and the open-system ages (Thompson et al., 2003; lower panel). $\sigma_{\delta^{234}U}$ is a measure for the variability of ($^{234}U/^{238}U$) in different sub-samples from an individual coral specimen. The numbers are used as plot symbols and denote the individual coral specimens listed in Table EA1. Only sub-samples that pass criteria (ii), (iii) and (iv) are shown. The rough trend between age variability and $\sigma_{\delta^{234}U}$ shows that specimens with a large variability in ($^{234}U/^{238}U$), which indicates a large variability in the degree of diagenetic alteration, also show a larger age variability. Because the Thompson et al. (2003) model does not correct the diagenetic effects (otherwise the trend should be removed in the lower panel), the coral specimens must have been altered by different diagenetic processes.

by the processes assumed by Thompson et al. (2003). The stricter test applied in this study reveals that other diagenetic processes are at least not negligible.

The results of this study demonstrate that the age variability, which is considered as an estimate of the true age uncertainty, of both the conventional and the open-system ages (Thompson et al., 2003) is larger than suggested by the quoted errors. Because both kinds of ages have been widely used for sea level reconstructions (Gallup et al., 2002; Cutler et al., 2003; Thompson and Goldstein, 2005; Frank et al., 2006; Scholz et al., 2006; Thompson and Goldstein, 2007) but also for calibration of the ^{14}C timescale (Bard et al., 1990; Chiu et al., 2005; Fairbanks et al., 2005; Chiu et al., 2006), it is an interesting question if the age variability depends on the coral age. In Fig. 9 the age variability is plotted against the mean sub-sample age for both the conventional (upper panel) and the open-system ages

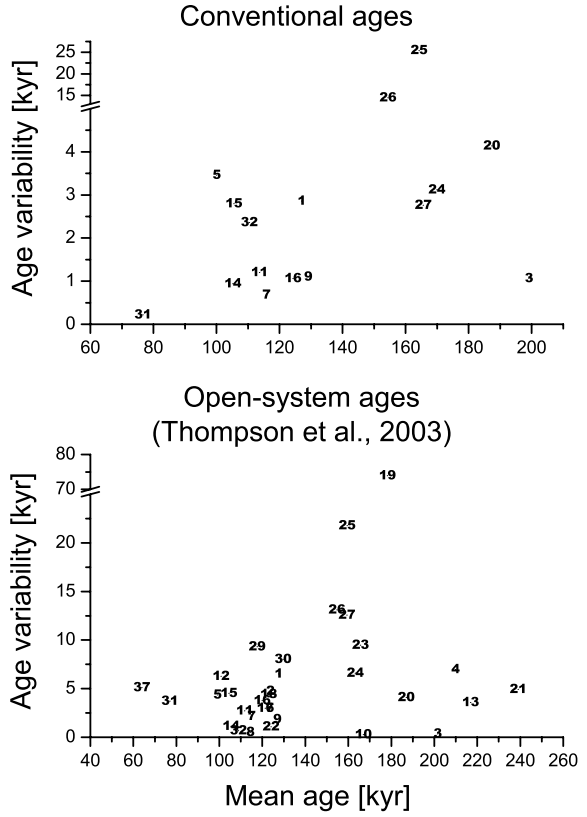

Fig. 9. Age variability vs. mean sub-sample age for both the conventional (upper panel) and the open-system ages (Thompson et al., 2003; lower panel). While the conventional ages seem to display a weak trend between age variability and age, the open-system ages (Thompson et al., 2003) do not show a trend.

(Thompson et al., 2003; lower panel). There seems to be a weak trend between the age variability of the conventional ages and coral age indicating that the age variability is enlarged with increasing age. This is reasonable because older samples most likely suffered a higher degree of diagenetic alteration. For the open-system ages (Thompson et al., 2003), no trend is visible. However, most specimens used in this study are from MIS 5, and the dataset contains only a few older specimens (i.e., MIS 6.5 and MIS 7). Holocene corals, which should be less diagenetically altered and, thus, have a smaller age variability, are not included. Therefore, this result may be considered as preliminary and should be confirmed by further studies using more data.

In summary, analysis of different sub-samples of an individual coral specimen has several advantages. For the conventional ages, the main advantage is the estimation of the true age variability, which can be compared to the quoted analytical errors. For the different model approaches, there are additional advantages (Scholz et al., 2006): (i) comparison of the measured and the predicted trend between ($^{234}U/^{238}U$) and ($^{230}Th/^{238}U$) and (ii) identification of diagenetic processes, which are not included in the models. In general, all models should only be applied to corals, which have been demonstrated to be altered by the underlying processes (Scholz and Mangini, 2007), and not to individual measurements from the literature (Thompson and Goldstein, 2005; Thompson and Goldstein, 2007). However, even if analysis of different coral sub-samples should give a more reliable estimate of the age uncertainty and, thus, represent an important advance in U-series dating of fossil corals, it is possible that all sub-samples of a specimen suffered diagenetic alteration, which is not detected by the widely applied criteria. In this case, the estimated age would significantly differ from the true age, and the age accuracy would be low. At present, the only possibility to solve this problem seems to be to apply combined $^{230}Th/U$- and $^{231}Pa/U$-dating to several sub-samples of one coral specimen.

6. CONCLUSIONS

The application of both conventional $^{230}Th/U$-dating and the different models (Thompson et al., 2003; Villemant and Feuillet, 2003; Scholz et al., 2004) to several sub-samples of an individual coral specimen reveals:

(1) The coral isochron age errors (Scholz et al., 2004) and the model age errors (Villemant and Feuillet, 2003) are substantially larger than the errors quoted for the conventional and the open-system ages (Thompson et al., 2003).
(2) The real age variability of the conventional ages, however, is substantially larger than suggested by the quoted errors (~350%).
(3) Similarly, the real variability of the open-system ages (Thompson et al., 2003) is substantially larger than suggested by the quoted errors (~400%).
(4) The widely used so called strict reliability criteria are not sufficient to detect all corals that were altered by other diagenetic processes than assumed by the models. Such samples can only be detected by analysis of several sub-samples of one coral specimen or combined $^{230}Th/U$- and $^{231}Pa/U$-dating.
(5) All models should generally only be applied to coral data, which have been demonstrated to be altered by the underlying processes. Application to published coral data (Thompson and Goldstein, 2005; Thompson and Goldstein, 2007) may result in both wrong ages and substantial underestimation of the age uncertainty.

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APPENDIX A. SUPPLEMENTARY DATA

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.gca.2007.01.016.

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