

## Pedogenic origin of dolomite in a basaltic weathering profile, Kohala Peninsula, Hawaii: Comment and Reply

### COMMENT

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Capo et al. (2000) addressed very important but unanswered questions about the origin of soil dolomite. In the article, Capo et al. examined the dolomites and claimed that the dolomites were of pedogenic origin. Here we claim that the dolomites are of diagenetic origin. The reasons are given in the following four points based on (1) crystallinity, (2) Sr isotopic value, (3) Mn/Sr and Fe/Sr ratios, and (4) the Ca/Sr ratio.

First, it is not hard to see in the X-ray diffraction (XRD) patterns of bulk powdered soil carbonate samples (Fig. 2 in Capo et al., 2000) that the soil carbonate samples they investigated are obviously poorly crystallized and impure. At  $2\theta \approx 20$ , there appears a peak in the four samples studied by Capo et al. that is significantly different from the peak of dolomite. Also there is a difference in the peak at  $2\theta \approx 52.5$  in sample no. P-2 and sample no. P-4 that is distinct from the dolomite peak. These XRD patterns may have resulted from the existence of clay minerals in the studied samples, which are consistent with the observation of Quade et al. (1995) that insoluble residues in soil carbonate samples account for 2%–60%.

Second, in most cases Sr isotopic ratios are different between the noncarbonate fraction and the carbonate fraction (Quade et al., 1995). At present, the principal method employed to determine the Sr isotopic composition of insoluble residue-containing carbonate samples is the one adopted at the Montanez Lab (Montanez et al., 2000). Clearly, if the Sr isotopic ratios in soil carbonate samples are directly determined, the values obtained would greatly deviate from the actual values of carbonate. Unfortunately, Capo et al. (2000) did not describe in detail the analytical method in their paper. As viewed from their simple description of the analytical method, we consider that the low Sr isotopic ratios they determined would not represent the real value of dolomite.

Third, in the study of the origin of marine carbonate, some authors made use of Mn/Sr ( $\leq 0.6$ ) and Fe/Sr ( $\leq 3$ ) ratios in carbonate to determine whether the samples have still preserved the original marine  $^{87}\text{Sr}/^{86}\text{Sr}$  records (Derry et al., 1994). Precisely, the major samples described in the paper of Capo et al. (2000) are in agreement with the above standards, but in sample no. P-4 Fe/Sr ratios are larger than 3. According to more strict differentiating standards (e.g., Mn/Sr  $\leq 0.1$  in dolomite; Montanez et al., 1996), sample nos. P-1 and P-4 would not be consistent with these standards, as is revealed by the occurrence of high-Mg calcite in sample P-1 and high contents of Fe, Mn, and Ba in sample P-4. Meanwhile, as can be seen in the Mn/Sr versus  $^{87}\text{Sr}/^{86}\text{Sr}$  diagram (Fig. 1), the samples described in the Capo et al. (2000) paper are of good linearity ( $R^2 = 0.88$ ). According to water-rock interaction modeling (Jacobsen and Kaufman, 1999), the above characteristics of the samples shown in Figure 1 may reflect the influence of interaction between primary dolomite and water. So we think that the samples examined by Capo et al. (2000), especially P-1 and P-4, have

been greatly reworked by postdiagenetic processes. For this reason, the Sr isotopic characteristics of the postdiagenetically reworked samples cannot provide clues to the provenance of cations in carbonate minerals.

Fourth, we claim that the dolomites should be considered to be of diagenetic origin rather than pedogenic origin. Based on the relationship between dolomites and primary fluids, i.e.,  $(\text{Sr}/\text{Ca})_{\text{dolo}} = D_{\text{Sr}}^{\text{dolo}} \times (\text{Sr}/\text{Ca})_{\text{fluid}}$  (see Budd, 1997), if we use published values of Sr and Ca, Sr = 700–740 ppm, Ca =  $2.17 \times 10^5$  ppm, and  $D_{\text{Sr}}^{\text{dolo}} = 0.012$ – $0.025$  (Vahrenkamp and Swart, 1990; Banner, 1995),  $(\text{Sr}/\text{Ca})_{\text{fluid}}$  ratios in the primary fluids responsible for pedogenic dolomites should be 0.129–0.284. It is known that the Sr/Ca ratio in meteoric water is 0.0046 (Banner and Hanson, 1990) and that in basalt is 0.0034–0.0055 (Quade et al., 1995). Obviously, meteoric water or soil-pore water derived from the former, if considered as the rock-forming fluids, could by no means produce such high-Sr dolomites as those described in the work by Capo et al. (2000). So we think that it is more reasonable to regard the dolomites examined in the work by Capo et al. (2000) as dolomites of diagenetic origin.

In summary, from the authors' samples, we claim that the majority of Sr and Ca in the dolomites was not derived from the basaltic parent material, and the dolomite was poorly crystallized and surely experienced diagenesis in a nonmarine environment.

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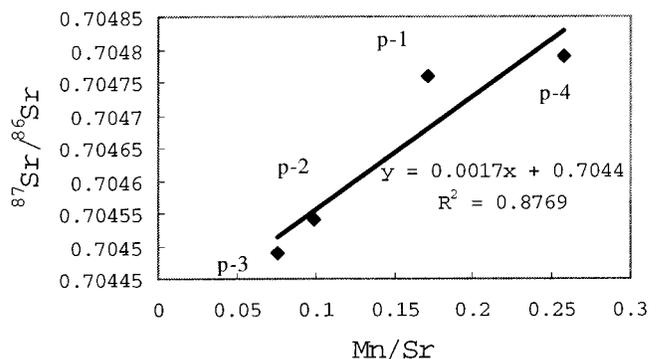


Figure 1. Mn/Sr vs.  $^{87}\text{Sr}/^{86}\text{Sr}$  plot (the numbers of samples and sample nos. are the same as in Capo et al., 2000).

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## REPLY

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Ji et al. use data from our article and from other work to argue that the dolomite we analyzed is not the result of soil forming processes. This conclusion is flawed because they (1) misinterpret relevant data and terminology, (2) do not consider the geologic context, and (3) do not fully understand soil carbonate formation.

**Terminology.** Their argument rests on the supposition that pedogenesis and diagenesis are distinct phenomena. They do not clearly define these terms; here we do, in order to address their arguments. Pedogenesis is the process of soil formation that occurs on land surfaces as atmospheric and biospheric acids interact with rock minerals to produce soluble and insoluble residues (e.g., Chadwick and Graham, 1999). Diagenesis refers to postdepositional processes, involving seawater, meteoric water, or subsurface brines, that alter sediments or sedimentary rocks up to the point of metamorphism, and include burial, compaction, dissolution, and precipitation (e.g., Morse and Mackenzie, 1990). The thermal regime and physicochemical processes encompassed by diagenesis and pedogenesis clearly overlap.

**Geological Context.** Ji et al. claim that the dolomites we described are “diagenetic” and were “greatly reworked by postdiagenetic processes” and not derived from the weathering of basalt. There is simply no field evidence to support a reworking hypothesis. The high permeability of the basalt flows that comprise Hawaii results in a water table near sea level, and subsidence and eustatic sea-level data show that our site has been >40 m above sea level throughout its history (see references within Capo et al., 2000). The carbonates were sampled from soil that has (1) never been buried by lava flows, (2) never been below sea level, or been influenced by ocean water or groundwater inundation, and (3) never incorporated marine sediment. The dolomite can only have formed in a soil environment, whether it precipitated as

a primary phase or was formed by the replacement of an earlier generation of pedogenic calcite.

**Pedogenic Carbonate Formation.** Carbonates are deposited in arid to subhumid soils when Ca and/or Mg, Fe, Na combine with  $\text{HCO}_3^-$  during soil desiccation, but subsequent wetting and drying episodes can lead to carbonate dissolution, equilibration with ambient conditions, and reprecipitation (McFadden et al., 1991). This affects crystallinity and geochemistry.

**Crystallinity.** Ji et al. state that our dolomite is “poorly crystallized” and “impure.” All pedogenic carbonates contain silicate minerals in an intimate physical mixture with their crystals (Chadwick et al., 1989). The small (<5  $\mu\text{m}$ ) size of the carbonate crystals and their intimate association with clay made it impossible to separate carbonate from other weathering products for X-ray diffraction (XRD) analysis, and accounts for the presence of other peaks in our samples. Regarding the crystallinity and stoichiometry of the dolomite crystals within the soil, we again point out (1) the presence of superstructure peaks d(015) and d(021) indicate our dolomite is well ordered (Fig. 2 of Capo et al., 2000), (2) molar Ca/Mg ranges from 0.9 to 1.0 in dolomite dissolved from the soil, and (3) SEM-EDAX (scanning electron microscope equipped with an EDAX energy dispersive system) analyses confirm a 50:50 Ca:Mg ratio in the dolomite.

**Trace element geochemistry.** Ji et al. use our data and a dolomite-water partition coefficient ( $D_{\text{Sr}}$ ) of ~0.02 to claim that Sr concentrations in our dolomites preclude their derivation from meteoric water or basalt. We note that other reported values are as high as 0.06 (e.g., Banner, 1995). Regardless, the  $D_{\text{Sr}}$  that Ji et al. use was determined by analysis of dolomite formed by diagenesis of marine calcite. The geological setting makes this comparison inappropriate. Further, trace element distribution coefficients are greatly dependent on precipitation rate, and rapid crystallization is associated with elevated Sr concentration in dolomite (>500 ppm; Bein and Land, 1983). We suggest that repeated wetting and drying episodes induced rapid crystallization and produced the high Sr concentrations (700–740 ppm) seen in our dolomites. We agree with Ji et al. that our Mn/Sr and Fe/Sr ratios are not consistent with well-preserved marine carbonate; these soil carbonates never existed in a marine environment.

**Sr isotope geochemistry.** The intimately mixed carbonate-silicate mineralogy of soil samples necessitates sequential leaching procedures to extract carbonate for analysis. Samples for isotopic and geochemical analysis of carbonate in our study were leached with ammonium acetate prior to dissolution in 2.0N acetic acid using methods described in detail in Capo et al. (1998); we regret that this fact was omitted in our article. At a profile subject to influx from sea spray, Whipkey et al. (2000) show that the  $^{87}\text{Sr}/^{86}\text{Sr}$  of the carbonate fraction is distinctly different from the silicate residue, with half the soil carbonate Sr derived from marine sources. In contrast, the  $^{87}\text{Sr}/^{86}\text{Sr}$  composition of ~0.705 for soil carbonate that we report is similar to that of the silicate residue (cf. Stewart et al., 2001), and indicates that over 90% of the carbonate Ca was derived from the weathering of basalt. The  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios show unequivocally that there is little to no marine component in the soil carbonate.

The dolomite we analyzed exhibits morphological, chemical, and isotopic characteristics produced by pedogenesis only. Further, its geological setting precludes all but a pedological origin. Direct use of marine-carbonate-derived distribution coefficients or trace element data is inappropriate. Capo et al. (2000) document well-ordered dolomite that results from soil-forming processes acting on basalt and tephra in an arid environment.

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## Multiple taxon–multiple locality approach to providing oxygen isotope evidence for warm-blooded theropod dinosaurs: Comment and Reply

### COMMENT

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Fricke and Rogers (2000) applied a unique approach to the question of theropod metabolism. Their premise was that latitudinal effects on the oxygen isotope composition of meteoric water should be reflected more directly in endothermic homeotherms, because they maintain constant body temperature, than in ectothermic heterotherms, because they will be colder at higher latitudes. The isotope slope of theropods appears steeper than that of crocodiles, and it is possible and perhaps probable that the interpretation of theropod endothermy is correct. However, we believe the authors were premature in dismissing environmental factors and sample bias as explanations for their data. We outline several of these factors to show how they may explain the data, but also to illustrate how Fricke and Rogers' study has further research implications.

**Thermoregulation.** Fricke and Rogers assume that crocodile body temperatures vary seasonally and latitudinally. However, all extant members of *Crocodylia* use several different strategies to maintain high body temperatures (25–35 °C), despite variations in ambient temperature. More critically, crocodiles only grow at body temperatures above a thermal minimum of ~25 °C (Coulson and Coulson, 1986) and will grow faster at warmer temperatures up to a thermal maximum of ~36 °C. Thus even if high-latitude crocodiles had mean annual body temperatures that were much cooler than equatorial crocodiles, their teeth probably grew at similar temperatures. We believe the theropod-crocodile isotope differences may reflect some factor(s) other than thermoregulation.

**Humidity.** Fricke and Rogers also assume that the body-water composition of crocodiles and theropods was the same at any given latitude or had the same slope across latitudes. This is not necessarily valid, because of behavior and the “humidity effect”:  $\delta^{18}\text{O}$  in terrestrial animals increases as humidity decreases (e.g., Luz et al., 1990). For example, suppose low-latitude crocodiles were aquatic during the day (low-humidity) and terrestrial during the night (high-humidity), similar to modern saltwater crocodiles, but higher latitude crocodiles were terrestrial during the day and aquatic at night, as are modern alligators. If so, then humidity effects will shallow the crocodile isotope slope relative to terrestrial theropods, even if body temperatures were

always identical. This effect would magnify if the crocodile diet differed taxonomically, so that low-latitude crocodiles ate only aquatic animals and high-latitude crocodiles also ate terrestrial vertebrates. Alternatively, suppose all crocodiles were fully aquatic with no humidity effects. The Scotese paleogeographic map for the Late Cretaceous shows that low-latitude sites were drier than high-latitude sites. If so, low-latitude theropods had an extra-high  $\delta^{18}\text{O}$  compared to their high-latitude cousins, steepening the theropod slope for reasons unrelated to thermoregulation.

**Seasonality.** Isotope compositions vary seasonally and are recorded in isotope zoning in tooth enamel (Fricke and O'Neil, 1996). In principle, isotope zoning studies on several teeth from a site can resolve the seasonal range of isotope compositions. However, in our experience, abundant fossil teeth of one species at a site often reflect one well-preserved specimen, not a random sample. Because the lifetime of a tooth is typically a few months to a year in crocodiles and in small dinosaurs (Erickson, 1996a, 1996b), the different teeth from each site may in fact represent only a single portion of the year. Homogenization of each tooth for analysis also must have blurred seasonality, which is a special problem for most sites with only 3–4 teeth. We believe that the mean tooth compositions may be biased toward one season, and insofar as seasons can vary by many per mil, that the crocodile-theropod isotope differences at most sites are not yet meaningful.

**Tooth growth.** Low-latitude tropical crocodiles may replace teeth all year, but high-latitude crocodiles probably will not if their body temperatures drop below the critical minimum. This will skew the isotopic data to higher values for the high-latitude crocodiles, whose teeth represent seasonal tooth deposition, further shallowing the crocodile isotope slope. In contrast, if theropods at high latitudes replaced their teeth all year, teeth representing the low  $\delta^{18}\text{O}$  winter-spring water will decrease the mean isotopic value relative to crocodiles. It would also create a greater standard deviation in the theropod data at higher latitudes, because some teeth would reflect winter deposition while none of the crocodile teeth would. For the Montana site, the isotope variability for theropods is much higher than for crocodiles. If isotope variability is a principal measure of metabolic physiology rather than behavior, then the Montana tooth data support the authors' original conclusion that theropods maintain a higher metabolic rate than crocodilians.

**Implications for paleoclimate analysis.** The results also have important implications for the study of paleoclimates, because gradients in meteoric water  $\delta^{18}\text{O}$  values reflect latitudinal heat distribution. For example, the data show a shallower  $\delta^{18}\text{O}$  slope (~0.2‰/lat) at

40–75°N latitude than is typical in the modern day (~0.3–0.4‰/lat). If the shallow Cretaceous slope is a global feature, it implies much more efficient poleward heat transport compared to today. The ability to use isotopes in fossil teeth to decipher the changes in the slope of the meteoric water line through time creates an important new means of investigating paleoclimate and heat distributions.

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#### REPLY

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We are pleased that our research has drawn the attention of others interested in applying stable isotope techniques to questions of paleobiology, and we are happy to reconsider our paper in light of the comments of Barrick and Kohn. However, we remain convinced that our interpretation of the data is the most parsimonious, and we maintain that  $\delta^{18}\text{O}$ -latitude gradients can be used to infer that some theropod dinosaurs were endothermic homeotherms. Lastly, we are glad that others are beginning to recognize the paleoclimatic implications of this research.

**Rationale.** Assessing behavior and physiology in the fossil record is a speculative endeavor at best. When faced with a staggering array of potential scenarios, we opted to minimize the number of assumptions and to focus on the simplest and most reasonable experimental designs. Our response is framed with this in mind.

**Thermoregulation.** Our primary assumption is that average crocodile body temperature varies with latitude. While behavior may allow crocodiles to modify body temperatures at any one location, there is no evidence that crocodiles living at different latitudes will act to maintain the exact same body temperature. In fact, it would not seem favorable to do so in the face of decreasing solar insolation and lower mean annual temperature at higher latitudes. This may explain why crocodiles are adapted to live and grow as their body temperatures vary over a 10 °C range. A more reasonable expectation is that crocodile body temperature decreases in a way that more or less mirrors latitudinal decreases in mean annual temperature even though body temperature is higher in each case due to behavioral strategies (e.g., mean annual temperature decreases from 25 to 15 °C, body temperature decreases from 35 to 25 °C).

The formation of tooth enamel over such a limited range of 35 to 25 °C can have a profound effect on  $\delta^{18}\text{O}$ , with a decrease in body temperature corresponding to increased isotopic fractionation of up to ~2.3% (Longinelli and Nuti, 1973). As a result, if body temperature decreases with latitude, the  $\delta^{18}\text{O}$ -latitude gradient of crocodiles should be significantly less than the gradient of a homeotherm characterized by a more uniform isotopic fractionation (see Fig. 1 of Fricke and Rogers, 2000). Therefore, we see no reason to readily dismiss the possibility that thermoregulation is the cause of different  $\delta^{18}\text{O}$ -latitude gradients between crocodiles and theropods.

**Humidity.** Humidity has an impact on  $\delta^{18}\text{O}$  of terrestrial animals because it modifies the  $\delta^{18}\text{O}$  of ingested water by increasing evaporation and shifting  $\delta^{18}\text{O}$  of water to higher values. Because evaporation occurs at the surface of leaves, this humidity effect can modify  $\delta^{18}\text{O}$  of herbivores that rely on leaf water to satisfy their water needs. Carnivores such as crocodiles and theropods do not generally eat leaves, so this humidity effect is expected to be much less, although they may inherit an effect by eating herbivores. The isotopic impact of humidity on  $\delta^{18}\text{O}$  of both herbivores and carnivores can be estimated using the physiological model of Kohn (1996). By changing the input value of relative humidity in this model from 50% to 90%, the predicted change in  $\delta^{18}\text{O}$  of an herbivore is 3.6‰, while the predicted change in a carnivore is only 0.2‰. This result indicates that a difference in humidity as transmitted via diet can have a large effect on herbivores (e.g. Luz et al., 1990), but is not likely to greatly effect  $\delta^{18}\text{O}$ -latitude gradients of carnivores.

The  $\delta^{18}\text{O}$  of surface waters of an entire region can be shifted to higher values relative to wetter locations as a result of increased evaporation. In this case, however, there is no reason to assume that only  $\delta^{18}\text{O}$  of theropods, and not coexisting crocodiles, would be affected. In fact, higher  $\delta^{18}\text{O}$  values of both crocodiles and theropods from the apparently more arid Madagascar locality relative to the more humid Texas locality support our argument, as was discussed in Fricke and Rogers (2000). Again thermoregulatory differences are the most parsimonious explanation of the differences in  $\delta^{18}\text{O}$ -latitude gradients.

**Seasonality.** We agree that the limited number of teeth analyzed from some of our localities is a weakness, but it is difficult to call on a seasonality sampling bias to explain different  $\delta^{18}\text{O}$ -latitude gradients for theropods and crocodiles. First, the samples already analyzed were derived primarily from vertebrate microsites, which tend to preserve many thousands of dissociated skeletal elements (including a plethora of teeth) in arguably time-averaged concentrations. The taphonomic characteristics of vertebrate microsites render it very unlikely that we repeatedly sampled single individuals. Second, a sampling bias explanation of the data requires that all crocodile teeth from high-latitude sites formed in summer and all coexisting theropod teeth formed in winter, while at low latitudes the reverse was true. While possible, this complicated scenario is also extremely unlikely. This discussion highlights the power of a method that requires an explanation of data obtained not from one single locality, but from many related localities.

**Tooth growth.** Although it requires assumptions regarding physiology and high-latitude seasonality, crocodile data from high latitudes could be skewed if tooth deposition occurred only during a warm season. Our data, however, are equivocal. The isotope variability for Montana crocodiles is indeed less than coexisting theropods, but for Alberta the isotopic variability is the same for both taxa (Fricke and Rogers, 2000). In either case, the conclusion is the same: different  $\delta^{18}\text{O}$ -latitude gradients do reflect differences in thermophysiology between taxa, and are not simply artifacts of behavior or sampling bias, as suggested by Barrick and Kohn.

**Implications for paleoclimate analysis.** A careful review of the literature reveals that we have already considered the paleoclimatic

implications of our stable isotope research (e.g., Fricke and Rogers, 1998; Fricke et al., 1998). However, it was clear to us that in order to estimate  $\delta^{18}\text{O}$  of Cretaceous precipitation, it was first necessary to resolve the nature of theropod thermoregulation. We feel that the data now available do provide reasonable evidence for theropod homeothermy, and we are in the process of preparing a more detailed report of how stable isotope data from theropods and other taxa can provide exciting new insights into Cretaceous climate.

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