Rare Earth Element Geochemistry and Taphonomy of Terrestrial Vertebrate Assemblages

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Most taphonomic analyses of vertebrate remains have focused upon physical processes. Chemical processes only rarely are addressed, leaving a large untapped store of quantitative taphonomic information contained within the bones themselves.

In this paper, the rare earth element (REE) signature of fossil bones in terrestrial deposits is shown to be controlled by the early diagenetic environment. Thus, bones fossilized in different early diagenetic environments may be separated by their distinct REE signatures. Furthermore, the variation of REE patterns developed in individual bones within an assemblage is controlled by sedimentologic and taphonomic processes. Hence, the degree of mixing and reworking (relative time and space averaging) of vertebrate elements within a particular assemblage may be determined from the REE patterns of the interred bones. REE geochemistry represents a new and powerful taphonomic tool.

INTRODUCTION

Morphological taphonomic analyses of vertebrate assemblages are extremely useful and, when combined with detailed sedimentological analyses, may help to determine the depositional histories and environmental settings of different vertebrate assemblages (e.g., Behrensmeyer, 1978; Fiorillo, 1988; Eberth, 1990; Behrensmeyer and Hook, 1992; Rogers, 1993; Smith, 1993). However, analyses based on surface features of bones are hampered severely by the large number of potential variables controlling bone-surface modifications in any one assemblage (Lyman, 1994; Cook, 1995). Because of these uncertainties, it is impossible, for instance, to identify individual reworked bones within vertebrate assemblages. Although this problem has been identified (e.g., Eaton et al., 1989), and addressed in specific environments (Lofgren et al., 1990), no satisfactory method exists to identify or measure reworking in most terrestrial assemblages. However, during the process of fossilization, bone incorporates elements from the early burial environment (Henderson et al., 1983). Accordingly, individual fossil bones (or suites of fossil bones within vertebrate assemblages) may permanently record fingerprints or signatures diagnostic of particular burial environments. If so, then bones that recrystallize in different depositional settings may inherit different trace-element compositions. The character and variation of these trace-element signatures then may be used to infer post-depositional transport and mixing within vertebrate assemblages, or to compare mixing and accumulation histories between assemblages.

Bone Fossilization

The inorganic (mineral) component of bone is calcium phosphate— $Ca_{10}(PO_4)_6(OH)_2$ Calcium phosphates form the vast majority of all vertebrate hard tissues (Young and Brown, 1982) and exhibit a wide range of physical and chemical properties. Bone is formed from hydroxyapatite (HAP) with a number of structural and chemical modifications. Common substitutions include: Na, Sr, Mg, and the rare earth elements substituting for Ca; CO_3 ,SiO₄ and HPO₄ substituting for PO₄; and F, Cl, and CO₃ substituting for OH.

Sedimentary (authigenic) apatite is rather stable in the sedimentary environment; however, bone mineral is soluble in seawater (Nriagu, 1983). The high reactivity of bone apatite occurs because bone crystals are very small, with a correspondingly high surface area (200 m²/g; Weiner and Price, 1986). Substitution of carbonate for phosphate in the apatite crystal lattice also causes distortions that further reduce the stability and crystallinity of the bone apatite crystals (Nelson, 1981; Nelson et al., 1983). During fossilization, these unstable, reactive, poorly crystalline materials alter to more stable, less reactive forms.

Fossil bone is normally composed of francolite (carbonate fluorapatite) which is the most stable form of apatite in sedimentary environments (Nathan and Sass, 1981). The recrystallization (fossilization) of bone is driven by the relative solubilities of biogenic apatite and sedimentary francolite in the burial environment. In essence then, bone 'fossilization' can be viewed as the process of recrystallization of reactive bone mineral to francolite, together with the loss of collagen and normally, but by no means exclusively, infilling of pore space with diagenetic minerals.

If a bone is to survive into the fossil record, the rate of recrystallization must exceed the rate of dissolution and destruction of bone mineral. Hedges et al. (1995) noted that archaeological bone generally is found either in 'well preserved' or 'very poorly preserved' categories, with few intermediate cases. This led to the suggestion that, once significant bone destruction occurs, the porosity of the bone is increased, and the surface area available for bone/ pore-water interactions and microbial activity also is increased. Thus, a positive feedback mechanism would be set up, leading to the rapid destruction of bone. This hypothesis is supported by the generally excellent histological preservation of fossil bone. The argument above suggests that the process of recrystallization occurs rapidly, during early diagenesis. Gillette (1993) remarked that Pleistocene mammal bones often tend to be soft and chalky, but that this texture is never found in bones older than 1.6 million years. He suggested that these chalky bones will never survive into the fossil record and, thus,

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preservation of bone must be complete within 1.6 million years, and probably very much earlier. Tuross et al. (1989) reported apatite crystal growth within 15 years after death. Certainly, growth of authigenic francolite can occur very rapidly, and is favored by conditions of organic decay. Briggs and Kear (1994) demonstrated that phosphatization of soft tissues can be initiated experimentally within a week, and probably occured even more rapidly in some cases.

In bone, however, the apatite crystals are protected by the protein matrix (Collins et al., 1995). This explains why bones do not dissolve rapidly on contact with seawater despite the high solubility of bone apatite crystals. The protective effect of the protein matrix has been demonstrated by heating bone powder. Bone apatite crystals alter on heating, but only after loss of the organic matrix (Person et al., 1995). Thus, while precipitation of francolite (and by analogy recrystallization of bone) may proceed rapidly, this process is probably slowed by the rate of collagen degradation.

A simplified model of bone fossilization can be produced where, under normal conditions, destruction of bone occurs as collagen is degraded and reactive bone crystals are exposed. Where burial conditions are favorable, phosphate concentrations build up in the localized environment of the bone, and bone apatite crystals 'recrystallize' to the chemically-stable francolite. This model emphasizes the importance of hydrology on bone fossilization, as leaching or biological recycling of phosphate will limit severely the preservation potential of the bone.

The precise mechanisms and rates of bone recrystallization are still poorly understood, and several possibilities exist: direct dissolution-recrystallization of apatite, overgrowth of francolite on biogenic apatite seed crystals, and reorganization of unstable crystals. The fidelity of histological preservation (including orientation of individual apatite crystals) argues against direct dissolution-recrystallization (Gillette, 1993; Zocco and Schwartz, 1994; Hubert et al., 1996; Schweitzer et al., 1997). It is difficult at present to distinguish between the seeding mechanism (Hubert et al., 1996) and processes of individual crystal reorganization. Both mechanisms result in increasing crystallinity and conversion of bulk mineralogy to francolite or fluorapatite. Hedges et al. (1995, p. 207) show that there is a relationship between crystallinity increase and microporosity decrease, noting that this relationship "is consistent with a processes by which smaller crystallites are either removed, or are "re-crystallized" to form thermodynamically more stable structures." Zocco and Schwartz (1994, p. 496) reported the preservation of original crystal alignment in Seismosaurus bone, but also stated that the electron diffraction patterns "match what would be expected from a calcium fluoride phosphate and francolite mixture", that the bone contains as much as 3.9 weight %fluorine, and that these mineralogies "are consistent with the common assumption that all fossil bone was originally dahllite (carbonate hydroxyapatite), changing to francolite (carbonate fluroapatite) with time." Other authors have suggested that crystallinity increases in bone may be caused by loss of carbonate from the lattice (Saliege et al., 1995; Person et al., 1995). All of these processes may contribute to crystallinity increases; however, dissolution of the smallest crystals alone cannot explain bone diagenesis, as the remaining crystals are usually either francolite or fluorapatite rather than the dahllite-like composition of biogenic apatite. Similarly, most fossil bones contain similar carbonate contents to modern bone (around 4–6 wt %). This shows that increases in the crystallinity of bone apatite during diagenesis are not caused by loss of structural carbonate from the apatite lattice. While the mechanism of bone recrystallization is unknown, it is clear that during fossilization extensive bone-groundwater interaction takes place. During this interaction, trace elements held in the surrounding waters may become incorporated into the crystal lattice of the fossil apatite. There is a large and growing body of literature documenting and describing the seemingly ubiquitous increase in trace-metal levels during bone recrystallization (e.g., Elderfield and Pagett, 1986; Wright et al., 1987; Williams, 1988; Grandjean and Albarède, 1989; Grandjean et al., 1993; Hubert et al., 1996; Denys et al., 1996; Holser, 1997); for instance, concentrations of the rare earth elements in fossil apatite from marine basins are higher than any other sedimentary mineral and commonly 5-6 orders of magnitude higher than seawater (Kolodny et al., 1996). Trace-metal increases are observed in both terrestrial and marine environments, and in archaeological and paleontological samples. These increases are not surprising, as apatite has a very strong affinity for a wide range of trace elements.

Stability of Early Diagenetic Trace Element Signals

The trace element signature developed in bone during early diagenetic recrystallization appears to be stable and resistant to later diagenetic change (Wright et al., 1987; Henderson et al., 1983; Williams, 1988). This argument has both theoretical and empirical support, and is vital to the arguments outlined below.

As discussed above, recrystallization of bone during early diagenesis results in a fossil bone composed of the relatively stable mineral francolite, with reduced porosity. The intracrystalline space is usually closed by growth of authigenic apatite (or other authigenic minerals), and the larger pore spaces are usually (but not always) infilled with other authigenic minerals (permineralization). Thus, the thermodynamic stability of bone crystals is increased, and the surface area of exposed apatite crystals is reduced. As porosity is reduced, and flow through the bone restricted, any further exchange of trace metals must proceed via solid-state diffusion. Several studies have demonstrated successfully that the trace element and isotopic composition of bone developed during early diagenesis is preserved throughout later diagenesis. For instance: Keto and Jacobsen (1987) were able to distinguish paleoceanic water masses on the basis of ϵ Nd signatures in Cambrian and Ordovician conodonts; Wright et al. (1987) used cerium anomalies in conodonts and ichthyoliths to reconstruct redox conditions in Paleozoic oceans; and Grandjean et al. (1987) used REE profiles in ichthyoliths to reconstruct paleodepth and changes in paleo-ocean circulation. Trueman and Benton (1997) showed that REE signals in bone survive reworking into contrasting sedimentary environments and, in the terrestrial realm, Williams et al. (1997) used REE patterns in Pleistocene bones to distinguish between oxidizing and reducing burial environments in Olduvai Gorge. Thus, the stability of early diagenetic trace element (REE) patterns has a strong empirical and theoretical basis. However, the initial incorporation of trace metals into bone will be governed by a number of factors, and does not necessarily reflect the trace metal composition of the environment of diagenesis.

Variables Controlling Diagenetic Trace-Element Enrichment in Fossil Bones

Fossil bones from the same skeleton (which might be assumed to have similar initial trace element contents, and to have experienced pore waters with similar compositions) show significant differences in their trace element content (Samiolov and Benjamini, 1996). This suggests that there are other variables affecting the trace element content of fossil bones. The controls affecting the final trace element composition of any exposed bone may be expressed as

$$\mathbf{X}_{i \text{ (bone final)}} = \mathbf{f} \left(\mathbf{X}_{i \text{ (bone initial)}}, \mathbf{X}_{i \text{ (pore water)}}, \mathbf{D}_{i}, \mathbf{K}, \mathbf{H}, \mathbf{M}, \mathbf{T} \right) \quad (1)$$

where X_i = concentration of trace element (i) in system X, D = apatite-fluid partition coefficient, K = chemistry of the microenvironment of burial, H = hydrology of the microenvironment of burial, M = bone microstructure, and T = length of exposure.

The first four variables on the left side of the equation effectively control the availability of ions for incorporation into apatite, whereas the last four effectively govern the rate of diagenesis of bone. Faster precipitation of minerals tends to force trace-element partition coefficients towards unity (Morse and Bender, 1990). Slower recrystallization tends to favor competition for lattice sites and increases the importance of mineralogical controls (Trueman, 1997).

Evidently, the incorporation of trace elements into bone during recrystallization in a terrestrial environment is a complicated process, with many variables determining both the absolute and relative concentration of trace metals present in the final recrystallized bone. Therefore, it is difficult to estimate original fluid (or bone) trace element compositions from fossil bone. However, the large number of potential variables means that the trace element composition of fossil bones is very sensitive to early diagenetic conditions.

RARE EARTH ELEMENTS

In this study, the rare earth elements (REE) are defined as the elements La (Z = 57) to Lu (Z = 71). The distinction between each element in the REE series is the number of 4f electrons. Since these 4f electrons are well screened from the nucleus by the completed 5s 5p sub-shells, they play almost no part in the valence forces. The shielding of the 4f electrons means that the addition of electrons cannot compensate for the increasing nuclear charge with atomic number. The REE form trivalent ions (with the exception of Eu and Ce); thus, the ionic radius of the REE³⁺ ion decreases smoothly as atomic number increases. This is known as the "lanthanide contraction" and, as the geochemical behavior of elements is governed to a large extent by the relative ionic radius and ion charge, the relative abundances of the REE in natural systems generally are controlled by ion size considerations. Many chemical processes fractionate the REE. Hence, plots of their relative abundances normally show smooth trends that can be used to infer the chemical processes responsible for this fractionation.



FIGURE 1—Schematic diagram illustrating fractionation of REE in terrestrial systems. Relative to the original REE source, soils are enriched in LREE, as the HREE are preferentially released to pore waters, and migrate from the weathering profile. Streams and rivers draining areas of REE weathering have lower La/Yb ratios than the soils they drain, and within rivers, the suspended and colloidal phase has a high La/Yb ratio compared to the dissolved phase (Goldstein and Jacobsen, 1988; Braun et al., 1993, 1998).

In this study, elements La to Nd are defined as light rare earth elements (LREE), elements Sm to Dy are the middle rare earth elements (MREE), and elements Tb to Lu are the heavy rare earth elements (HREE). The concentration of REE in geological systems usually is plotted relative to an international standard. A chondrite standard is used normally in igneous systems as an approximation of bulk earth concentrations, whereas an average shale standard is used normally in sedimentary or aqueous studies as an approximation of average upper crustal compositions. In this study, the North American Shale Composite (NASC) values of Gromet et al. (1984) are used to normalize raw data.

During terrestrial weathering the LREE are preferentially adsorbed onto mineral surfaces and remain in the weathering profile, whereas the HREE form more stable aqueous (carbonate) complexes and preferentially are removed into solution (e.g., Roaldset, 1973; Duddy, 1980; Wood, 1990; Morey and Stetterholm, 1997; Braun et al., 1993, 1998). Thus, in a single fluvial system, pore waters associated with floodplains and soils may be depleted in HREE compared to pore waters associated with river channels (Fig. 1). The extent of this fractionation will vary according to the chemistry and hydrology of the fluvial system.

REE in Bones

The REE apparently reside in the two calcium sites in the apatite lattice, and are normally present in living bone at the ppb level (Arrhenius et al., 1957; Shaw and Wasserburg, 1985). Analyses of fossil bones yield high REE levels, commonly in the 10³ ppm range (Kolodny et al., 1996). Pore-water REE compositions vary, but are commonly below 1 ppb. Thus, the REE are concentrated into bone during diagenesis (Henderson et al., 1983; Wright et al., 1987). This led Kolodny et al. (1996) to suggest that the presence of REE in a fossil bone indicates that it has undergone early diagenetic alteration.

The REE composition of ichthyoliths collected from the modern ocean floor is enriched with respect to the living tissue and 5-6 orders of magnitude higher than seawater, but has a REE pattern and isotopic composition similar to the overlying bottom water. This led Wright et al. (1987) to suggest that ichthyoliths are enriched in REE while at the sediment-water interface, and that the REE are not fractionated during this process (however, Reynard et al. (1999) argue convincingly for fractionation of REE between seawater and ichthyoliths). If the REE are not fractionated between seawater and apatite, or if fractionation of REE is relatively minor, then the ichthyolith faithfully records the bottom water REE signature. This has allowed the reconstruction of paleo-redox conditions and patterns of ocean circulation from fossil ichthyoliths and conodonts. This is possible because of the relatively stable chemistry of oceanic bottom waters, and the long residence time of ichthyoliths at the sediment-water interface. The more complex diagenetic system operating in terrestrial environments means that all bones in a single assemblage may not (and do not) necessarily inherit the same REE pattern.

While the trace element content of individual bones may be related to the pore-water chemistry in the early burial environment, the *variation* in REE concentrations of equivalent bones within an assemblage is controlled by sedimentologic and taphonomic variables

$$V_{(as)} = f(V_{(b,e)}, R_{(d)})$$
 (2)

Where $V_{(as)}$ is the variation in trace element content of bones within an assemblage, $V_{(b,e)}$ is the variation in the original burial environments experienced by the bones sampled, and $R_{(d)}$ is the rate of introduction of bones into the final deposit (related to the rate of sediment deposition and extent of reworking—essentially the amount of time bones spend in different burial environments).

From these controls we can make two predictions:

- (1) Assemblages with more rapid rates of accumulation/ reworking (i.e., low time averaging) should show less variation in trace element patterns, as the bones will be concentrated rapidly in similar depositional environments.
- (2) Assemblages with similar reworking rates should show greater variation if the source area for the bones is a more complex, varied environment (linked to space averaging).

While many studies have reported variations in apatite REE composition through a time-sequence, relatively few have documented variation in REE composition within individual terrestrial assemblages.

CASE STUDIES

Two terrestrial fluvial vertebrate-bearing deposits were selected to determine the variation in REE composition in bones within and between assemblages. The Two Medicine Formation of the Willow Creek Anticline, north-west



FIGURE 2—Map showing sampled locations and regional outcrop of the Judith River Group, and Two Medicine Formations (from Eberth and Hamblin, 1993).

Montana, USA, and the Dinosaur Park Formation of Dinosaur Provincial Park, Alberta, Canada (Fig. 2), were chosen as they are broadly contemporaneous (Campanian), contain a similar fauna in terms of bone size and structure, and represent different taphonomic environments. The primary REE source is similar in both formations, the REE being derived from contemporaneous acidic volcanism related to the growth of the Rocky Mountains, and possibly derived from a single source, the Elkhorn Mountain volcanic complex (Thomas et al., 1990).

Two Medicine Formation

Lorenz and Gavin (1984) separated the Two Medicine Formation of the Willow Creek Anticline section into four sub-facies—the lower, northern, lake, and upper sub-facies. The lake and northern sub-facies are described as contemporaneous and laterally distinct. The lower sub-facies is characterized by medium- to fine-grained sand bodies interpreted as low sinuosity, channel-confined stream and crevasse-splay deposits, and well-developed calcareous paleosols, set within red and green silty muds. Vertebrate remains are commonly found as isolated scattered bones.

In contrast, the northern sub-facies is characterized by medium to coarse-grained sand bodies with well-developed scour features, and purple and green silty muds. The sand bodies are interpreted as higher energy, poorly confined channel flow and sheet floods. The red muds and well-developed paleosols found in the lower subfacies are absent, and the sediments of the northern sub-facies have a lower carbonate content (Varricchio et al., 1999). Vertebrate remains commonly are found as isolated bones throughout the northern subfacies, although associated and occasionally articulated material is also found.

Near the top of the lower sub-facies (within the northern sub-facies of Lorenz and Gavin, 1984) is a conspicuous bone bed, which can be traced throughout the Willow Creek Anticline section (Horner and Gorman, 1988; Varricchio and Horner, 1993). The bed contains numerous disarticulated and fragmented dinosaur bones. The bone density varies between locations, with the maximum being around 4,500 bones in a 70-m² area (Horner and Gorman, 1988). Average bone concentrations, however, are around 30 bone fragments per square meter (Varricchio and Horner, 1993). The matrix is a black, silty, calcareous mud with a high ash content (Horner and Gorman, 1988).

The bone bed is interpreted as a debris flow on the basis of its lateral extent, the three-dimensional orientation of bones within the matrix, the erosive base, and the abundance of reworked caliche clasts and rip-up mud-balls. The inconsistent thickness of the bone bed also is indicative of deposition related to a debris flow, flowing over irregular topography. Varricchio and Horner (1993) came to the same conclusion.

Like most bone beds elsewhere in the Two Medicine Formation, the bone bed from the Willow Creek Anticline section has a low taxonomic diversity (Varricchio and Horner, 1993; Rogers, 1993) and has been interpreted as a mass-death assemblage. Horner and Gorman (1988) noted that the bones are very poorly preseved and fragmented, and they terminate in transverse, clean fractures. Hooker (1987) suggested that the bone bed formed by reworking of permineralized remains in a debris flow, possibly as a result of a breached lake. However, Schmitz et al. (1998) suggest that the majority of bones are, in fact, relatively complete, and were introduced into the bone bed as fresh, unpermineralized remains. The process responsible for the concentration of bones is unclear. The bone bed may contain the concentrated, time-averaged remains of bones reworked from lower deposits into the debris flow (and therefore reflect an attritional deposit). In this case, low taxonomic diversity reflects the dominance of a particular taxon in the environment over a period of time equivalent to the time averaging of the deposit. Alternatively, the bone bed may represent the remains of a catastrophic death event, bones being reworked into the bone bed either relatively quickly (unpermineralized) or after permineralization.

Samples from the lower and northern subfacies were taken from fresh weathering surfaces within overbankmud, channel-sand, and paleosol horizons. No attempt was made to sample extensively from individual beds, except from the major bone bed, because of the difficulty of correlation across the sample area. Bones were sampled from two bone bed quarries-the 'Children's Dig' and 'Make-a-Wish' sites.

Dinosaur Park Formation

Dinosaur Provincial Park contains extensive three-dimensional exposures of the uppermost 90 m of Campanian sediments of Judith River age. Eberth and Hamblin (1993) revised the stratigraphic status of the Judith River Formation and elevated it to the Judith River Group, includ-

Age Ma Montana Alberta DPF Two Medicine Formation 75 Lithofacies CAMPANIAN Upper 76 Oldman Fm. 77 ithofacies Foremost 78 Middle Fm. 79 FIGURE 3-Summary stratigraphy, and tentative correlation of the

N-W.

S.

Two Medicine Formation and Judith River Group in northwestern Montana and southern Alberta. Samples in this study were taken from the lower portions of the Dinosaur Park Formation and the upper lithofacies of the Two Medicine Formation. Stippled region indicates the approximate position of the Willow Creek Anticline section, and the bold line represents the bone bed (based on: Lorenz and Gavin, 1984; Eberth and Hamblin, 1993; and Rogers et al., 1993).

ing the Foremost, Oldman, and Dinosaur Park Formations (Fig. 3). The upper portions of the Oldman Formation and the Dinosaur Park Formation are exposed in Dinosaur Provincial Park.

The Oldman Formation is lithologically and temporally equivalent to part of the Two Medicine Formation of Montana and may be traced throughout northern Montana and southern Alberta (e.g., Eberth and Hamblin, 1993, fig. 2). The Dinosaur Park Formation forms a southward-thinning clastic tongue that reaches its greatest thickness (approximately 120 m) near Edmonton, Alberta. The age of the Dinosaur Park Formation has been determined by ⁴⁰Ar/³⁹Ar and K-Ar dating of bentonites from within 5 m of the basal contact of the Dinosaur Park Formation and 12 m of the top of the formation (Eberth and Hamblin, 1993). These have yielded dates of 76 (± 0.5) and 74 (± 0.5) Ma., respectively. The sediments of Dinosaur Provincial Park have received a great deal of attention, and the sedimentology, taphonomy, and paleoenvironment are very well constrained (e.g., Dodson, 1971, 1973; Thomas et al., 1987; Wood et al., 1988; Brinkman, 1990; Eberth, 1990; Eberth and Hamblin, 1993).

In contrast to the Two Medicine and Oldman Formations, the Dinosaur Park Formation is dominated by fineto medium-grained sandstone units. The sand:mud ratio is higher-approximately 3.15 (70% sand), calculated from logs in Eberth and Hamblin (1993). The sands form a number of facies associations, mainly deposited within meandering river channels. Wood et al. (1988) calculated channel depths of 7–25 m and widths of 55 to >120 m. Eberth (1990) and Eberth and Hamblin (1993) concurred with this interpretation. In addition, Eberth (1996) noted the occurrence of tidally-influenced, mud-filled channels at the top of the sequence.

Vertebrate Accumulations in the Dinosaur Park Formation

Most vertebrate remains within the Dinosaur Park Formation occur as scattered bone accumulations either as channel-lag traction-concentrated accumulations (Wood et al., 1988; Eberth, 1990) or as accumulations concentrated on lateral accretion surfaces (Wood et al., 1988). Articulated skeletons are relatively common and occur within lateral accretion units, channel lags, and overbank units. Samples were taken from two main fossil assemblages within the Dinosaur Park Formation—BB 20 and BB 41.

BB 41 and it's lateral equivalent BB41a are situated towards the eastern end of the park, on the north side of the Red Deer River. These are located towards the base of the Dinosaur Park Formation. The bone bed is an overbank deposit (Getty et al., 1998), and is overlain by a sand unit with a silt component containing fragmented plant debris. BB41 and BB41a are monogeneric (>90% of identifiable remains are assigned to Centrosaurus sp.; Getty et al., 1998). These monogeneric ceratopsian bone beds are thought to represent catastrophic mortality events, possibly the death of a collection of ceratopsian dinosaurs in a single flood or storm event. However, all bones recovered from BB 41 (and some other ceratopsian bone beds in the Dinosaur Park Formation) are disarticulated and commonly show traces of abrasion, polish, and fragmentation, which reflect a period of exposure between deposition and burial (Getty et al., 1998). Most of the fractures seen are characteristic of fresh breaks, and none of the bone remains show surface modifications characteristic of extensive subaerial weathering on modern bones (Getty et al., 1998).

BB 20 is located stratigraphically in the central portions of the Dinosaur Park Formation, on the west side of the park, on the south bank of the Red Deer River. The bone bed is contained within fine-to-medium sands with largescale, low-angle cross stratification typical of point-bar lateral accretion deposits (Wood et al., 1988). The bone bed is associated with small *in situ* ironstone nodules, and the bones are iron-stained. BB 20 is interpreted as a lag deposit. It is multi-generic and contains abundant, mildly abraded, and disarticulated bone fragments.

ANALYTICAL METHODS

Sample Collection

High rates of erosion in both sampled localities ensure that many fresh fossil bones can be found on weathered slopes throughout both areas. The vertebrate material collected from these assemblages was isolated from the matrix and required little or no preparation. Bones were sampled from either fresh weathering surfaces or directly from active quarries. Since the REE do not appear to be physiologically vital trace elements, and as *in vivo* bone concentrations are several orders of magnitude lower than diagenetic concentrations, the REE record is taxon-independent. Thus, indeterminate bone fragments may be used for destructive chemical analysis. In most cases, compact cortical bone fragments were collected. Identification of taxonomic affinity was uncertain and was not attempted.

Sample Preparation and Analysis

ICP-MS Analysis

The REE content of bone samples was measured by ICP-MS (inductively coupled plasma mass spectrometry) analysis at the University of Bristol. Prior to analysis, the external surfaces of bone (the outermost 10 mm) were mechanically removed to avoid the effects of recent weathering. Any adhering sediment was removed mechanically with an engravers drill. The bone sample was then washed with distilled water and placed in an ultrasonic tank to remove any further adhering sediment. Bones were ground to powder with an agate mortar and pestle, and placed in clean glass sample vessels. Prior to further preparation, all samples were heated to 100°C for 24 hours to remove water.

Dried powders of bone and sediment samples (0.2g)were digested at 200°C with 69% Primar HNO₃ in Teflon containers. Samples were evaporated on a hotplate until the sample became a nearly dry cake. After cooling, the cake was dissolved with ~ 15 ml of 5% HNO₃ on a hot plate, made up to 100 ml with 1% HNO₃ in a volumetric flask, and stored in a clean plastic bottle until analysis. Most samples dissolved completely with this treatment, however a few samples contained residual silicate solids. In common with other pore-filling authigenic minerals, these residual silicate solids contain very low REE concentrations, and do not significantly alter the REE pattern of the whole bone. Silicate minerals may be dissolved in HF, however adding HF to calcium-rich solutions produces insoluble calcium fluoride precipitates, which may remove REE from solution.

REE measurements were performed on a VG Plasma Quad 2+ mass spectrometer. The ICP-MS operated in scanning mode between masses 100.9 and 189.8, using the pulse counter, and the following isotopes were measured: ¹⁰²Ru, ¹³⁹La, ¹⁴⁰Ce, ¹⁴¹Pr, ¹⁴⁶Nd, ¹⁴⁷Sm, ¹⁵³Eu, ¹⁵⁵Gd, ¹⁵⁹Tb, ¹⁶²Dy, ¹⁶⁵Ho, ¹⁶⁶Er, ¹⁶⁹Tm, ¹⁷⁴Yb, ¹⁷⁵Lu, and ¹⁸⁷Re. Rhenium and ruthenium were used as internal standards and were kept at a constant concentration of 100 ppb for all samples and standards. Calibration was carried out using REE standards at concentrations of 0, 20, 60 and 100 ppb. All samples were run in triplicate, and at least three international rock standards and blank samples were analyzed during the course of each batch of analyses to monitor accuracy and precision. Estimated errors (2σ) are below ±10% in all cases.

While ICP-MS analysis provides a rapid and relatively reliable method to analyze all REE simultaneously, problems exist when measuring REE concentrations in rocks with relatively high concentrations of barium. This is because Ba forms oxides that potentially interfere with many of the REE (Greaves et al., 1989). However, the major interference exists with europium. Thus, samples with relatively low REE concentrations, and relatively high Ba concentrations, may show positive Eu anomalies. Ba also forms isobaric overlaps with ¹³⁹La. Hence, samples with anomalously high La and Eu contents may be identified as possibly showing Ba interference. In a larger study of 250 bones, including the bones described in this study, 18 bones were shown to suffer from Ba interference (Trueman, 1997). Barite can be detected by XRD analysis in all but two of these samples. No other samples contained detectable barite. However, to avoid potential unrecognized errors, Pr was used in place of La to represent the LREE, and no inferences were drawn from either Ce or Eu anomalies.

XRD Analysis

The mineralogy of bone samples was determined by Xray diffractrometry (XRD). Dried powders of bone and sediment were mounted onto glass discs by mobilization with acetone. XRD spectra were obtained at the University of Bristol on a Philips PW1800 diffractometer. Spectra were obtained with a copper anode, operating at 45 kV and 40 mA. Full spectra were measured from 6° 2 θ to 70° 2 θ , with a step size of 0.02° 2 θ and step time of 2 seconds per step.

RESULTS

All fossil bone samples yield XRD patterns consistent with francolite (carbonate fluorapatite). Secondary calcite, quartz, iron oxide, barite, and clay minerals were also detected in some bones. REE concentrations in bones and sediment are presented in Appendix Tables 1A and 1B.

Total REE Contents

The total REE contents measured in bones range from 16–9,300 ppm (mean 1136) in the Two Medicine Formation, and from >30–4,600 ppm (mean 1544) in the Dinosaur Park Formation (Appendix Tables 1A, 1B). These levels are similar to total REE levels recorded previously in ichthyoliths and conodont remains (mean total REE concentrations from 124 ichthyoliths = 922 ppm—Grandjean et al., 1993; Girard and Alberède, 1996).

The mean REE contents of sediments (73 ppm and 100 ppm from the Dinosaur Park Formation and Two Medicine Formation, respectively) from the two formations are significantly lower than bone REE contents and are similar to average sediment REE concentrations (Appendix Tables 1A, 1B).

The total REE concentrations within mixed mineral samples depend upon the relative amounts of each mineral phase present in the sample and the REE concentration in each mineral phase. Apatite, with its very high affinity for the REEs, frequently contains at least two to three orders of magnitude higher REE concentrations than any other mineral phase present in the bone samples. While the REE patterns of mixed mineral samples are relatively insensitive to the non-apatite mineral content, the total REE concentrations partly will reflect the relative apatite content in the bone samples. However, Hubert et al. (1996) report Ce and La values in Jurassic dinosaur bones ranging from 100-1400 ppm and 100-1000 ppm, respectively. These values were obtained by direct analysis of apatite by electron microprobe. While the spot size of the microprobe is larger than the inidividual crystal size, Hubert et al. (1996) demonstrate that apatite was the only major mineral phase analvsed. Thus, their REE totals do not reflect varying apatite/authigenic mineral ratios, but directly demonstrate the variation of REE concentrations in the apatite of fossil bones from terrestrial environments. Apatite/calcite ratios were approximated crudely from XRD patterns as the maximum counts for the major apatite (c. 2.79 Å) and calcite (c.

FIGURE 4—Frequency histogram comparing the range and distribution of shale-normalized Pr/Yb ratios in bones from the Dinosaur Park Formation (DPF) and the Two Medicine Formation (TMF). Note that the bones from the Two Medicine Formation are more varied than bones from the Dinosaur Park Formation and that approximately half of the bones from the Two Medicine Formation are LREE-enriched. NASC values of Gromet et al. (1984) are used for normalization, and subscript -n refers to shale-normalized values.

3.04 Å) peaks, and in a smaller set of samples, by weight loss with reaction with dilute acetic acid. Apatite/calite ratios did not correlate with total REE contents in either case. Analysis of the acetic acid soluble fraction of the bone powder (i.e., authigenic calcite) shows that the REE content of the permineralizing calcite is 2–3 orders of magnitude lower than the apatite.

REE Patterns

Bones from both assemblages yield a variety of shalenormalized REE patterns. These patterns can be expressed simply in terms of the ratios of light to middle and heavy rare earth elements (e.g., Sm_n/Pr_n; Sm_n/Yb_n, and Pr_n/Yb_n ratios, where n indicates shale-normalized concentrations). With one exception, all bones analyzed from the Dinosaur Park Formation are HREE-enriched, whereas bones from the Two Medicine Formation have a much greater range in Pr/Yb ratios, approximately half being LREE-enriched (Fig. 4). The Two Medicine Formation assemblage is significantly more varied in terms of fossil bone REE patterns than the Dinosaur Park assemblage (99% level F-test based on Sm/Yb ratios, Table 1,) and, within individual assemblages, the bone bed shows significantly less variation than the other Two Medicine Formation assemblages (95% level F-test based on Sm/Yb ratios, Table 1).

DISCUSSION

Very high total REE concentrations (>7000 ppm) have been determined in previous studies of terrestrial bones (e.g., Tauson et al., 1991; Denys et al., 1996; Samoilov and Benjamini, 1996). Hubert et al. (1996) relate the trace-element composition of dinosaur bones from the Brushy Ba-



TABLE 1—Variance of LOG(Pr/Yb)_N ratios of individual bones within terrestrial vertebrate assemblages, and significance values (F-Test) of differences in variance between assemblages (N.S. = no significant difference, DPF = pooled sample of all Dinosaur Park Formation bones, TMF = pooled sample of all Two Medicine Formation bones). LOG values of (Pr/Yb)_N ratios used to ensure normal distribution of values. Note that attritional Two Medicine Formation assemblages are significantly more varied than all Dinosaur Park Formation assemblages. Also note that, within the Two Medicine Formation, the bone-bed assemblage is significantly less varied than the lower and northern subfacies assemblages (95%, 99% levels, respectively). Within the Dinosaur Park Formation, BB20 is significantly more varied than BB41.

							LSF	
	BB41	BB20	DPP	LSF	BB	NSF	$^+$ NSF	TMF
Variance	0.03	0.12	0.10	0.5	0.07	0.28	0.38	0.31
BB41 BB20 DPP	$\begin{array}{c} 0.01\\ 0.01 \end{array}$	0.01 N.S.	0.01 N.S.	$\begin{array}{c} 0.01 \\ 0.01 \\ 0.01 \end{array}$	0.05 N.S. N.S.	0.01 N.S. 0.05	$\begin{array}{c} 0.01 \\ 0.01 \\ 0.01 \end{array}$	$\begin{array}{c} 0.01 \\ 0.05 \\ 0.01 \end{array}$
LSF BB NSF LSF + NSF TMF	$\begin{array}{c} 0.01 \\ 0.05 \\ 0.01 \\ 0.01 \\ 0.01 \end{array}$	0.01 N.S. 0.05 0.01 0.05	0.01 N.S. 0.01 0.01 0.01	0.01 N.S. N.S. N.S.	$0.01 \\ 0.01 \\ 0.01 \\ 0.01$	N.S. 0.01 N.S. N.S.	N.S. 0.01 N.S. N.S.	N.S. 0.01 N.S. N.S.

sin Member of the Morrison Formation to proximal, contemporaneous volcanism. They explain the high levels of trace elements (including REEs) found in fossil bones to the breakdown of silicic ash within the formation and, hence, to high pore-water trace-element concentrations. This model suggests that assemblages with a higher volcaniclastic content should show greater total REE enrichment in autochthonous bone samples. In this study, however, the mean total REE concentration of bones from the Two Medicine Formation is significantly lower than the mean total REE concentration in bone samples from Mongolian sediments (c. 7000 ppm, Samoilov and Benjamini, 1996) with a much lower volcaniclastic content (Jerzykiewicz et al., 1993). This suggests that breakdown of volcaniclastic material within the burial environment is not responsible solely for the high REE concentrations seen in some terrestrial environments.

Does the REE Content of Fossil Bone Reflect the Environment of Burial?

Many authors have noted that recent and fossil ichthyoliths record REE patterns similar to the overlying seawater. Thus, the fossil bone or tooth may inherit an unfractionated record of the overlying sea water. However, the extent to which bones from terrestrial environments inherit an environmental signal is currently unknown.

Terrestrial weathering appears to fractionate the REE, the HREE being preferentially removed into solution, whereas the LREE remain within the weathering profile (Fig. 1). Thus, if bones do inherit the REE pattern of the pore waters in the early burial environment, then bones hosted within clay-rich overbank or soil environments may possess higher Pr/Yb ratios than bones hosted within channel sands (however, the extent of weathering-related REE fractionation depends on the chemistry of the fluvial system). Bones from the overbank-dominated northern



FIGURE 5—Ratios of light (Pr) to heavy (Yb) REE concentrations in bones from terrestrial assemblages. Note that bones from the two assemblages sampled from Dinosaur Provincial Park (BB 20 and BB 41) are consistently more HREE-enriched than bones from the northern sub-facies of the Two Medicine Formation. Furthermore, within the northern sub-facies, bones sampled from sandstone channels are more HREE-enriched then bones sampled from mudstones and lie within the field of the Dinosaur Park Formation. Bones from BB20 are also slightly more HREE-enriched than bones from BB41.

sub-facies of the Two Medicine Formation are indeed consistently more LREE enriched than bones from the channel dominated Dinosaur Park Formation (Fig. 5). Furthermore, within the northern sub-facies of the Two Medicine Formation, bones recovered from course grained sandstone beds clearly are distinct from those recovered from overbank deposits (Fig. 5) and are strongly HREE-enriched. Similar (although less pronounced) distinctions are seen between the channel-hosted assemblage BB20, and the overbank-floodplain-hosted BB41 from the Dinosaur Park Formation. Bones from channel sand facies are consistently more HREE-enriched than bones from overbank, clay-rich facies. Thus, it appears that in terrestrial environments (as in marine environments) the final REE pattern developed in equivalent fossil bones may reflect the early burial environment. A similar conclusion was reached by Hubert et al. (1996) who stated that the chemistry of dinosaur bones reflects the composition of the ground water. However, this does not mean that bones in either terrestrial or marine settings possess a totally unfractionated record of the pore-water REE composition.

Another possible explanation for the differences in REE patterns between the two assemblages could be differences in the non-apatite fractions of the bone samples. The ratio of partition coefficients (La/Yb) between aqueous REE (as carbonate complexes) and calcite is approximately 63 (Zhong and Mucci, 1995), whereas the ratio of partition coefficients (La/Lu) between aqueous REE (as carbonate complexes) and hydroxyapatite is approximately 5.1 (Koeppenkastrop and De Carlo, 1992). Thus, as calcite preferentially incorporates LREE and fractionates the REE to a greater extent than apatite, a greater proportion of calcite in the bone sample could increase the observed LREE enrichment. However, because of the lower absolute values of all partition coefficients between aqueous REE and calcite compared to apatite, extremely low apatite/calcite ratios would be needed to alter the bulk REE patterns significantly. Analysis of calcite separated from bones from the Two Medicine Formation show very low to-



FIGURE 6—Variation in REE patterns in vertebrate assemblages from terrestrial and marine environments. Coastal marine sample includes fish and reptile bones from a Late Triassic (Rhaetian) bone bed at Aust Cliff, Gloucestershire, England, and from an Early Cretaceous bone bed at Durlston Bay, Dorset, England (values are given in Trueman, 1997). Open marine samples consist of condont remains from the Devonian Coumiac Limestone of Southern France (Girard and Albarède, 1996). Note that the field of variation described by bones from terrestrial environments is much larger than that of either marine environment (significant at the 99% level; F-test), and that the field described by bones from the Two Medicine Formation is also larger than that described by the Dinosaur Park Formation (99% significance, F-test).

tal REE levels, with no strong LREE enrichment (Trueman, 1997). Therefore, the differences in REE concentrations between bones from the Two Medicine Formation and Dinosaur Park Formation most likely are related to differences in the sedimentary burial environment between the two assemblages.

Variation in Trace Element Patterns in Vertebrate Assemblages

The variation in REE composition in bones in an assemblage was predicted to be linked to the amount of mixing (time and/or space averaging) of bones within the assemblage. This can be tested by comparing the REE variation in assemblages of bones from environments with uniform early burial environments (e.g., open marine basins) to the variation in bones from environments with highly varied early burial environments (e.g., terrestrial fluvial assemblages). The rate of final burial is faster in terrestrial assemblages, and this will tend to lower the variation in REE values found in terrestrial assemblages compared to marine assembalges; however, bones from the deep marine environment are still significantly less varied (1% F



FIGURE 7—Variation in REE patterns in bones from three assemblages from the Willow Creek Anticline section of the Two Medicine Formation. The bone bed may contain reworked bones from either the lower or northern sub-facies, or it may represent a collection of relatively fresh bones. Note that the field described by bones from the bone bed is contained entirely within the field of the lower sub-facies, but not within that of the northern sub-facies. Note also that the variation of REE composition in bones from the bone bed is significantly lower than either the lower or northern sub-facies (99% significance, respectively). This suggests that the bone bed represents a restricted sample (limited time averaging), but reworking from a restricted source within the lower sub-facies cannot be ruled out.

test) than bones from terrestrial fluvial settings (Fig. 6). The increased variation in terrestrial assemblages is controlled by the complexity of bone-groundwater interactions in the terrestrial environment compared to the buffered marine environment.

Time Averaging

The rate of final burial is a measure of time averaging. Therefore, if two assemblages can be matched in terms of early burial environments, the trace element chemistry of the bones can be used as a relative measure of time averaging between the two assemblages. By comparing the variation in trace element composition between two assemblages, inferences can made concerning the degree of mixing of vertebrate remains, either in terms of the variation in original burial environments, or the relative time averaging.

Isolated, disarticulted bones from the Dinosaur Park Formation show significantly less variation in terms of their REE patterns than bones from attritional assemblages from the Two Medicine Formation (Fig. 6, Table 1). If one makes the assumption that the potential range of available early burial environments was similar in both fluvial environments, this suggests that during deposition of the Dinosaur Park Formation, bones were rapidly introduced into fluvial channels (and, thus, experienced similar physical and chemical conditions of early diagenesis). In contrast, during deposition of the Two Medicine Formation, bones remained in a variety of floodplain/soil/channel-sand environments. Thus, the REE chemistry of disarticulated remains from the two formations indicates that time averaging prior to introduction into fluvial channels was relatively low in the Dinosaur Park Formation (i.e., bones were concentrated rapidly into fluvial channels). Vertebrate remains in the Two Medicine Formation are dominated by scattered, disarticulated remains, with few articulated skeletons. REE chemistry indicates that the bones remained in many different depositional settings throughout diagenesis.

REE Variation in Bone Beds: Provenancing and Depositional Controls

Two Medicine Formation Bone Bed

The main bone bed sampled from the Two Medicine Formation is situated near the transitional boundary between the lower and northern sub-facies. As bones from these two subfacies have contrasting REE profiles, it is possible to test the amount of reworking present in bones from the bone bed (Fig. 7).

If the bones were introduced by extensive reworking of prefossilized lower sub-facies deposits, one would expect the geochemistry of bones from the bone bed and the lower sub-facies to be similar. If, however, the bone bed sampled only bones from the northern sub-facies, then the geochemistry of bones in the bed should resemble those from the northern sub-facies. Alternatively, the bone bed may have sampled material from a unique burial environment, in which case the bones from the bed may not correspond to either the northern sub-facies or the lower sub-facies. If the time interval between deposition of the bones and introduction into the bone bed was relatively short, then variation in the bone bed will be relatively low compared to the total variation in the lower and northern subfacies.

The REE composition of the bones from the bone bed fall entirely within the range of bones from the lower sub-facies and partially overlap the range of bones from the northern sub-facies (Fig. 7). The variation in bones from the bone bed is significantly less than either of the other sub-facies (Table 1). None of the bones sampled from the bone bed fall in the range of bones sampled from channel sands from the northern sub-facies. Thus, the REE evidence shows that the bone bed represents a restricted sample, either reworking bones from a narrow range of burial environments or incorporating relatively fresh bones (low time averaging). Sedimentological evidence shows that the bone bed was deposited as an erosive debris flow, potentially cutting into a wide range of different burial environments. If the majority of bones were reworked after early diagenetic recrystallization in these diverse depositional environments, they would be expected to have a wide range of compositions. Accordingly, it appears that the bones were introduced while they were relatively fresh, rather than as pre-fossilized bones. Thus, the bone bed probably represents a deposit with relatively short time averaging.

Dinosaur Park Formation Bone Beds

A similar approach can be taken in analyzing bone beds from the Dinosaur Park Formation. Sample site BB 41 is a strong candidate for interpretation as a catastrophic bone accumulation. It is, as far as can be ascertained, monogeneric, and appears to represent a single flood event that reworked bones exposed for a relatively short period of time (Getty, Eberth pers. comm., 1998). Throughout the Dinosaur Park Formation there are a number of similar, monogeneric, usually ceratopsian, bone beds, similarly interpreted as catastrophic flood events. BB20 contains a mixed, disarticulated assemblage of bones from a variety of taxonomic groups contained within lateral accretion units or channel lags in a large sand-dominated channel system. Both bone beds contain disarticulated and mildly abraded remains, and it is difficult to tell whether differences in taxonomic diversity of the bone beds reflect taphonomic differences (rate of accumulation, time averaging) or paleoecologic differences (low diversity faunal associations).

Earlier it was predicted that increasing time and space averaging within bone beds should increase the variation in trace element composition in the preserved bones. As these two assemblages are relatively close geographically and stratigraphically, it is reasonable to suggest that the range of available early depositional environments was similar for both deposits. Bones from BB20 display significantly more variation than bones from BB41, showing that BB41 is less time averaged than BB20. However, BB20 is significantly less varied than all Two Medicine Formations with the exception of the bone bed (Table 1). Thus, if one accepts the assumption that the potential range of available early burial environments was similar in both fluvial environments, these results allow for the comparison of relative degrees of time averaging in vertebrate assemblages both within and between formations.

CONCLUSIONS

The trace element concentration of an *individual* bone is a function of a number of interrelated variables that may be summarized as those factors controlling the trace element content and availability in the pore water (e.g., source rock, weathering rates, water content, partitioning of trace elements between fluid and mineral phases), and those factors that control the rate and style of apatite recrystallization (e.g., hydrology and chemistry of burial environment, mineral suite present, bone microstructure and porosity, microbial activity). The results of this study suggest that the REE composition of individual vertebrate remains reflect the environment of early diagenesis in terrestrial as well as marine settings. If this is true, then a method exists to separate reworked from non-reworked elements within mixed assemblages, to provenance individual bones within mixed assemblages, and to reconstruct aspects of the early diagenetic environment. Furthermore, the variation in REE content within and between assemblages can be used as a relative measure of time and/or space averaging. The potential of this method is limited by the rate at which diagenetic apatite forms in fossilizing bones and the variability of pore-water REE concentrations in different environments. This limitation may be lessened by extending the method to other environmentally sensitive trace metals. The results suggest that bone bed assemblages within the Dinosaur Park Formation of Dinosaur Provincial Park are less time-averaged than assemblages from the Two Medicine Formation. The Two Medicine Formation of the Willow Creek Anticline section itself contains both time-averaged attritional assemblages, and a low-diversity bone bed with relatively limited time-averaging.

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ACCEPTED JUNE 29, 1999

Sample	Lithology	La	Ce	\mathbf{Pr}	PN	Sm	Eu	Gd	$_{\mathrm{Tb}}$	Dy	Ho	Er	Tm	Yb	Lu
E5	LSF mud	56.82	45.03	4.60	17.37	3.50	2.13	8.14	1.30	11.27	3.55	9.94	1.46	8.02	1.13
E6	LSF mud	151.79	264.00	32.64	131.88	30.61	8.91	41.73	4.78	25.77	5.25	9.89	0.93	4.78	0.66
E37 E80	LSF mud	5.47	6.06	0.77	3.51	0.70	0.55	0.80	0.18	1.17	0.39	1.22	0.21	1.41	0.22
E33	LSF mud	00 297	190.44 063 60	20.72	121.65 164 87	20.00 58.18	10.6 86.66	105 90	0.40 95 18	11.05 124.88	04.14 0.114	14.88 71 74	1.61 6.83	8.90 26 89	1.33 3.69
E52	LSF mud	97.85	53.31	3.19	9.83	1.17	0.53	7.42	0.42	3.90	1.25	5.39	0.93	6.95	1.28
E53	LSF mud	285.04	443.31	43.40	191.26	20.32	7.67	36.52	8.46	40.21	6.47	18.20	2.09	8.85	1.15
E60	LSF sand	30.95	14.84	1.20	3.80	0.53	0.41	7.33	0.11	1.18	0.47	2.84	0.68	6.18	1.54
E61	LSF mud	12.22	2.18	0.10	0.26	0.07	0.28	3.90	0.00	0.27	0.14	0.87	0.21	2.57	0.55
E62	LSF mud	9.45	4.16	0.35	1.21	0.34	0.14	1.96	0.07	1.08	0.44	2.05	0.37	2.57	0.40
E63	LSF mud	140.94	214.24	23.67	90.40	18.06	6.22	38.19	4.72	30.57	6.86	18.49	2.16	10.71	1.62
E64 E65	LSF sand	1319.39	1839.89 7 80	182.77	622.64 9.11	109.91	28.84 0.15	166.32	17.77	100.31	19.47	48.25	5.25	23.79	3.03
E02	LSF sand	4.4.1	9.89	0.98	2.11	0.43	GL.U	1.33	GU.U	0.38	0.08	0.27	0.02	0.00	0.04
El	BB mud	153.06	125.46	12.48	39.27	6.34	1.95	9.84	1.33	9.45	2.22	5.92	0.77	4.84	0.81
E2	BB mud	28.86	21.21	2.37	77.7	1.09	1.18	2.08	0.21	1.93	0.54	1.60	0.31	2.20	0.39
E3	BB mud	90.29	65.27	8.75	29.16	5.47	1.80	6.69	0.94	6.31	1.66	4.93	0.75	4.64	0.68
E21 E40	DD mud	563.20 96 80	775.92	81.60	327.71	60.64 1.04	18.13	59.51 1.20	10.01	52.14 1 oc	8.84	24.28	2.95	16.43 0.20	2.56
E40 E41		00.00 2 00	09.02 4 10	4.10 0 50	14.21 1 60	1.34 0.00	0.00	1.09 0.09	0.06	00'T	0.43	0.30	0.04	0.09	0.03
Е41 Е44	BP mud	0.02 902 64	4.10 226.70	0.00	108 25	07.01 16.70	00.0 79.6	0.00 17 85	0.00	07.0	9.78	01.U	1.02	0.10 5 95	10.0
E.45	BR mind	65.43	79.50	7.35	97.59	5 05	5.58	98 8	0.79	4.47	1 19	9.64 2.64	0.37	0.37	0.35
E46	BB mud	31.05	21.84	2.16	11.56	1.94	31.04	1.89	0.24	1.50	0.37	1.10	0.23	1.91	0.43
E47	BB mud	49.34	42.04	6.48	24.57	3.32	0.68	2.51	0.42	2.03	0.59	1.25	0.15	0.72	0.08
E48	BB mud	8.39	7.59	0.86	4.29	0.89	15.51	0.73	0.13	0.63	0.10	0.31	0.04	0.30	0.04
E49	BB mud	596.35	654.93	65.23	263.10	45.94	8.69	58.63	8.28	46.58	11.16	39.93	4.20	27.23	3.99
E50	BB mud	47.67	46.65	4.52	16.97	2.43	1.54	3.00	0.43	2.52	0.65	1.66	0.24	1.58	0.25
E7	NSF sand	12.88	21.63	2.43	9.23	1.56	0.53	1.63	0.20	1.22	0.28	0.70	0.12	0.72	0.12
E8	NSF mud	771.81	1327.14	99.81	335.97	59.41	17.66	87.08	10.21	60.67	14.03	38.82	5.39	32.67	4.98
E9	NSF mud	857.54	1271.22	121.25	419.43	72.29	19.94	97.48	10.01	61.11	13.02	33.83	4.12	24.58	3.59
E10	NSF mud	356.58	556.87	80.17	307.75	60.83	17.60	69.25	7.94	41.53	7.15	15.83	1.57	7.86	0.97
EII	NSF mud	141.97	189.78	20.77	80.43	16.26	10.11	22.16	2.78	17.08	3.79	10.52	1.34	8.61	1.35
E24 P05	NGF mud	400.09 70.06	110.00	00.00 12 71	240.10 75.00	12.14	20.02	04.00 12.04	0.00	30.14 10 <i>56</i>	1.92	20.19 7 44	2.79	16.0T	2.00
E26	NSF mud	7.88	69 °C	0.92 D	1.52	0.30	10.50	10.03 0.23	11.2	0.32	1.34 0.09	4.44	0.05	0.40	0.11
E27	NSF mud	150.59	157.01	17.32	64.92	10.61	11.13	12.35	1.79	9.64	1.95	5.17	0.68	4.20	0.62
E28	NSF mud	541.92	967.76	127.22	560.08	114.59	19.91	96.24	15.50	77.38	13.94	31.37	3.45	20.87	2.51
E30	NSF mud	556.16	843.73	123.15	533.28	104.98	18.00	68.28	14.08	65.64	13.46	26.85	3.07	14.78	2.03
E31	NSF mud	4.30	1.90	0.27	1.65	0.49	15.30	0.21	0.05	0.26	0.06	0.17	0.02	0.22	0.06
E32	NSF mud	6.28	3.43	0.37	1.45 1 7 4	0.40	9.08	0.33	0.0 0.04	0.48	0.12	0.44	0.12	0.69	0.12
500 F 26	NSF sand	0.00 59 30	0.00 70 5.4	7 1 7	1.14 95 03	0.44 7.65	166 166	16.0	9.01	0.23	0.03 6 34	0.75 90.75	0.03	98 09	0.00 1 8.7
E54	NSF mud	1501.34	2236.79	339.81	1353.83	271.90	72.75	460.26	54.44	301.42	54.52	144.18	16.06	81.98	10.62
E55	NSF mud	1647.62	3518.67	463.50	1978.22	417.61	114.31	583.34	62.18	302.49	49.10	122.83	11.81	54.73	6.58
E57	NSF mud	926.88	1105.51	124.45	486.59	90.91	24.64	197.69	19.02	114.38	24.23	76.57	9.52	53.83	7.95
E66	NSF sand	31.00	28.21	2.50	9.68	2.00	0.70	7.33	0.78	7.38	2.47	10.05	1.80	12.16	2.21
E67	NSF sand	159.29	523.95	29.10	123.65	30.53	8.27	54.14	10.33	89.51	25.97	97.90	16.85	111.53	20.63
Sediment															
	LSF mud	20.73	44.86	4.82	20.99	4.74	1.32	3.84	0.90	4.20	0.28	0.70	0.12	0.72	0.12
	BB 100	15.44	30.99	3.93	16.44	3.46	1.00	3.31	0.46	2.62	0.58	1.64	0.24	2.02	0.28
	NSF sand	12.88	21.63 29 95	2.43 6 90	9.23	1.56 1 EO	0.03	1.63 1 21	0.20	1.22 20.0	0.65	1.79 1	0.27	1.64 9.01	0.26
	NSF mud	35.36	02.20 03.01	0.20	04.02 44.00	8.00 8.95	ео.т 1 26	4.01 6 90	1 90	07.0 8 45	0.07	0 70	0.40	10.2	0.19
	Bentonite	0.39	1 01	0.08	0.65	0.18	0.06 0.06	0.31	0.06	0.52	0.65	1 78	0.97	1.64	0.26
ļ		2000		0000	0000	0	0000	-	0000		0000				0

KEE concentration	ns (ppm) u	n bones and	sequment		a i incouti		.11010							
Sample	La	Ce	\mathbf{Pr}	PN	Sm	Eu	Gd	$^{\mathrm{Tb}}$	Dy	Ho	Er	Tm	$_{\mathrm{Yb}}$	Lu
BB20.1	137.01	197.52	20.36	100.95	14.37	5.86	30.90	5.46	34.49	7.86	27.10	4.26	24.41	3.08
BB20.2	266.08	418.53	50.55	245.64	57.34	17.63	101.13	14.62	104.63	25.65	88.52	11.88	73.51	12.78
BB20.3	169.03	244.13	28.75	136.26	31.36	8.97	61.55	8.09	58.84	15.02	52.93	6.77	41.38	7.31
BB20.4	833.92	1274.95	213.29	955.91	240.73	68.87	375.54	53.15	330.24	72.03	221.25	27.64	158.84	24.68
BB20.6	361.31	485.86	54.99	254.24	55.68	17.70	108.90	14.25	98.75	24.22	80.50	10.30	64.30	11.18
BB20.7	13.78	17.25	1.94	9.13	2.04	1.49	17.24	0.80	7.77	2.77	13.39	2.35	18.27	3.74
BB20.8	26.04	24.65	2.82	14.59	3.29	1.39	12.05	1.61	15.78	5.32	23.42	3.57	25.25	4.87
BB20.9	454.08	596.73	68.42	319.25	70.30	20.27	127.19	17.20	124.31	30.86	108.80	13.97	86.94	14.94
BB20.10	1066.59	1515.12	174.07	875.36	245.20	60.36	637.16	45.83	401.31	118.83	386.51	27.90	329.34	50.04
BB20.11	503.49	652.97	77.15	372.67	85.68	28.88	173.21	26.13	198.27	50.17	175.45	22.94	140.73	23.16
BB20.12	423.30	572.78	67.31	314.67	75.20	25.29	173.24	22.76	155.95	38.04	123.68	15.46	90.94	14.40
BB20.13	771.07	1179.68	190.02	848.07	203.73	62.30	335.55	46.15	303.36	69.34	220.63	28.07	160.25	24.56
BB20.14	191.38	205.27	21.49	98.09	20.90	6.98	61.20	5.92	43.23	21.11	45.37	6.58	44.40	8.13
BB20.15	127.33	97.99	10.32	59.38	11.81	5.90	42.32	11.09	73.24	19.57	81.29	17.60	58.89	9.98
BB41.A1	81.67	118.81	12.36	56.02	8.83	3.70	21.74	4.49	22.82	4.38	16.17	2.05	13.56	1.47
BB41.A6	74.73	171.97	20.31	108.49	27.58	9.10	31.08	5.13	45.68	8.19	22.85	3.12	20.80	
BB41.A8	14.19	32.93	4.53	22.82	6.18	2.40	7.83	0.62	4.73	1.04	3.54	0.54	3.28	
BB41.A9	162.07	371.29	45.54	187.89	49.06	13.33	79.40	10.97	81.31	15.27	46.72	6.42	27.19	
BB41.A10	373.06	636.17	79.58	347.09	81.35	23.20	141.88	18.60	122.43	27.89	89.74	11.44	67.20	10.24
BB41.A11	91.05	176.27	19.69	95.45	18.92	5.68	28.84	6.52	35.94	60.9	20.25	2.83	16.76	1.93
BB41.A13	5.64	7.97	0.79	3.33	0.53	0.59	8.52	0.13	0.74	0.13	0.45	0.05	0.22	0.05
BB41.A14	84.85	71.20	6.21	24.37	4.03	1.11	10.29	0.93	6.70	1.68	6.20	0.86	5.75	1.01
BB41.12	29.91	35.14	3.57	14.74	2.85	0.87	7.28	0.62	4.51	1.24	5.03	0.77	5.88	1.17
BB41.13	129.49	166.44	17.85	73.18	14.43	5.22	41.85	4.07	26.70	6.41	20.78	2.60	15.30	2.49
BB41.14	314.24	368.10	36.95	138.23	24.37	7.23	56.09	5.18	32.22	7.44	24.11	3.01	17.78	2.97
BB41.15	362.94	517.10	58.52	268.32	68.78	16.81	162.64	10.94	81.73	23.07	69.95	4.39	50.89	8.10
BB41.16	760.74	1113.54	162.37	705.08	168.74	53.07	305.50	41.65	262.28	57.55	178.87	22.10	127.36	19.90
BB41.17	384.90	519.97	56.20	252.10	44.41	18.68	91.07	21.14	119.71	25.89	98.33	21.50	65.62	10.72
BB41.18	1087.52	1528.69	158.39	701.61	119.77	48.03	201.74	47.37	230.74	44.96	153.34	33.24	95.35	14.84
BB41.19	11.60	12.69	2.03	10.52	3.85	0.92	4.96	0.71	2.35	0.54	2.43	0.36	2.28	0.39
BB41.20	309.39	281.51	24.86	92.80	14.97	5.05	51.81	4.24	30.55	8.46	30.68	4.19	27.07	4.72
BB41.21	553.54	770.15	78.37	353.53	43.51	20.36	92.99	22.14	135.42	26.91	92.70	11.14	71.21	7.71
BB41.22	274.30	318.93	31.79	131.79	25.39	8.19	63.40	6.67	44.83	10.59	34.88	4.53	26.51	4.32
BB41.23	423.26	502.87	54.15	219.15	44.32	14.12	99.49	11.74	79.63	19.47	63.11	8.05	47.60	7.44
Sediment														
BB20.Sand	18.70	30.89	3.53	18.99	3.70	1.62	5.31	1.21	4.10	0.98	3.22	0.62	3.68	0.51
BB41.Nodule	1.60	2.87	0.59	2.63	1.17	0.49	1.09	0.28	0.85	0.19	1.04	0.18	0.00	0.21
BB41.Sand	19.35	41.17	4.80	21.27	5.42	1.96	5.44	1.27	4.17	1.11	2.90	0.72	0.00	0.61

APPENDIX II