STABLE ISOTOPE GEOCHEMISTRY OF KAOLINITE FROM THE "WHITE SECTION," BLACK RIDGE, CLERMONT, CENTRAL QUEENSLAND: IMPLICATIONS FOR THE AGE AND ORIGIN OF THE "WHITE SECTION"

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Abstract—Kaolinite from the Black Ridge, Clermont, has relatively low δ^{18} O (12.3‰ to 14.8‰) and very low δ D values with a large variation (-120‰ to -85‰). Comparison of these data with those from the nearby Denison Trough and elsewhere in eastern Australia, together with previous studies of the mineralogy of the sedimentary rocks, suggests that extensive kaolinization of the "White Section" resulted from weathering during the Late Triassic to Early Jurassic periods. The relatively large variation in δ D values of kaolinite probably derives from post-formational isotopic exchange with other fluids.

The similarity between δ^{18} O values of kaolinites from Black Ridge and from the Denison Trough suggests that the small Miclere-Black Ridge basin may have been part of the Denison Trough before the Late-Triassic inversion. The preservation of original δ D values in kaolinite at Black Ridge indicates that unlike the Denison Trough, which was reburied at more than 1000 m, the Miclere-Black Ridge basin was not reburied at great depth during the Mesozoic period.

Key Words-Inversion, Isotopic exchange, Kaolinite, Late Triassic, Low &D, Weathering.

INTRODUCTION

A number of studies, beginning with the work of Savin and Epstein (1970), have shown that kaolinite forms in isotopic equilibrium with its environment. δD and δ^{18} O data for kaolinites from different clay-rich soils all plot very close to the "kaolinite line" (Lawrence and Taylor, 1971, 1972), implying that the kaolinites were in isotopic equilibrium with coexisting meteoric water. A stable isotope study of kaolinite from the Santa Rita Deposit indicates that there is a clear distinction in stable isotope data between shallow supergene and deep hypogene kaolinites (Taylor, 1974, 1979). Recently, some stable isotopic studies on kaolinite have been focused on interpreting its age and formation conditions in eastern Australia (Bird and Chivas, 1988, 1989; Baker and Golding, 1992) as there has been an increase in the δD and $\delta^{18}O$ values of the surface meteoric waters since the Late Paleozoic as continental drift moved Australia from high to low latitudes resulting in a progressively warmer climate (Bird and Chivas, 1988).

The kaolinite in this study is from an extremely altered pebbly sandstone known as the "White Section" in a Permian basin in the Clermont area, central Queensland. The "White Section" was first reported by Reid (1936), but the age and origin of both the sandstone itself and the kaolinite alteration have remained controversial. Its contact with the coal measures is described as an unconformity by Osman (1971), and a possible Tertiary age was suggested by Osman and Wilson (1975). A Permian age has also been proposed for the sandstone itself, as *Glossopteris* has been recognized in the unit, but the intensive kaolinization is regarded as the result of either diagenesis or leaching, probably during the Tertiary (Beeston, 1978). A late Permian age is further suggested by a 5 m bed of kaolinitic sandstone found stratigraphically higher than the late Permian "Clarkei Bed," which is a regionally persistent marine horizon in the western Bowen Basin (Cook and Taylor, 1979). It has even been proposed that the term "White Section" should not be used and that this unit be simply regarded as an oxidized section of the Blair Athol Coal Measures (Preston, 1985).

In the present study kaolinites from the "White Section" have been analyzed for their D/H and ¹⁸O/¹⁶O ratios. Although the age of the last isotopic equilibration is uncertain, when these data are placed in the context of other stable isotopic analyses of kaolinite and the geological constraints, they are significant for understanding the age and formation condition of the kaolinite and, to some extent, the tectonic evolution of the Permian basins at Clermont.

GEOLOGICAL SETTING AND CHARACTERISTICS OF THE "WHITE SECTION"

The Clermont gold field is approximately 335 km northwest of Rockhampton, Queensland. The goldfield is situated at the eastern margin of the Anakie Inlier and borders the Bowen Basin to the east and the Drum-

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Figure 1. Regional geology of central Queensland (modified from Malone, 1964; Olgers, 1969a, 1969b). Legend: 1 =Permian sediments in the Clermont gold field; 2 = Permo-Triassic sediments; 3 = Upper Devonian granite; 4 = Middle Devonian volcanics; 5 = Proterozoic(?) Anakie metamorphics.

mond Basin to the southwest (Figure 1). The goldfield comprises a series of NNW-trending semi-isolated early Permian basins, which are tectonically aligned with the Denison Trough. The development of these basins is directly related to the evolution of the Denison Trough (Dickins and Malone, 1973).

The Black Ridge gold deposit is located in the north of the Clermont goldfield (Figure 1). The stratigraphic units in the deposits are the Anakie Metamorphics (Proterozoic?), which are composed of chlorite-muscovite schists with minor quartzite; the early Permian Blair Athol Coal Measures, including up to 50 m of basal conglomerates and up to 45 m of an upper unit of conglomerate with minor shales and coal seams; the "White Section," composed of up to 24 m of pebbly kaolinitic sandstone; and Tertiary basalts.

The "White Section" is well developed in the northern part of the Clermont goldfield. At Black Ridge, it conformably overlies the Blair Athol Coal Measures, unconformably overlies the Anakie Metamorphics, and is overlain by oxidized Tertiary basalt or black soil. The "White Section" is normally 5 to 15 m thick, but a thickness of up to 24 m has been recorded in the Miclere–Black Ridge basin (Zhou, 1992). Most of this rock unit is strongly oxidized or decomposed and comprises a pale, pebbly sandstone with an extensively kaolinized matrix to up to 30 vol. % kaolinite in some specimens, as confirmed by XRD analysis.

In thin sections, the remaining pebbles are composed mainly of quartz, since lithic and feldspathic pebbles



Figure 2. A) Coarse kaolinite and fine kaolinite: a = coarse kaolinite; b = fine kaolinite. B) Fine kaolinite around coarse kaolinite, indicating two generations of kaolinite: a = coarse kaolinite; b = fine kaolinite. Length of field of view is 400 μm .

have completely decomposed and been replaced by kaolinite and minor, fine muscovite or illite. Rare remnant ferruginous bands are observed at the bottom of the rock unit, which probably represent oxidized siderite. Most of the kaolinite occurs in grains $\leq 2 \mu m$, but a small proportion (<10%) is $\geq 5 \mu m$, and much better crystallized. The coarser kaolinite is often surrounded by the finer one (Figure 2), indicating its earlier formation.

SAMPLES AND METHOLOGY

Five kaolinite samples were collected from chips from percussion drill holes. Kaolinite separation is conventionally regarded as difficult, especially in the presence of other clay minerals. For this work, the separation method presented by Longstaffe (1984) was modified slightly, resulting in separates of >92% purity as confirmed by both XRD analysis and microscopic observation (Zhou, 1992).

The samples were analyzed for oxygen and hydrogen isotopic ratios in the Isotope Geochemistry Laboratory, University of Queensland. The oxygen and hydrogen compositions were determined using the stan-

Table 1. Stable isotopic data for kaolinite from the "White Section," Black Ridge.

Sample no.	Latitude	Longitude	δ ^{ι8} O‰	δD‰	Depth (m)
k2	22°37′S	147°33'E	13.0	-120	-8
304	22°37′S	147°33'E	14.8	-111	-8
k l	22°37′S	147°33'E	14.5	-104	-12
207	22°37′S	147°33'E	13.8	-96	-14
309	22°37′S	143°33'E	12.3	-85	-18

dard techniques detailed by Clayton and Mayeda (1963) and Coleman *et al.* (1982), on a VG602E mass spectrometer. The results of the stable isotopic analyses are reported in δ notation relative to the SMOW standard for both oxygen and hydrogen; analytical uncertainties are $\pm 0.2\%$ for δ^{18} O and $\pm 3\%$ for δ D values.

BASIC EQUATIONS FOR THE STABLE ISOTOPIC STUDY

The following equations were used for the calculation and assessment of the relationship between δD and $\delta^{18}O$ values of meteoric waters and the stable isotopic fractionations between kaolinite and water (all in per mil).

The "meteoric line" from Craig (1961) is:

$$\delta D_w = 8 \cdot \delta^{18} O_w + 10 \tag{1}$$

The "kaolinite line" calculated from data of Savin and Epstein (1970) is:

$$\delta \mathbf{D}_{\mathbf{kao}} = 8 \cdot \delta^{18} \mathbf{O}_{\mathbf{kao}} - 224 \tag{2}$$

The equation for oxygen isotopic fractionation between kaolinite and water was taken from Land and Dutton (1978):

1000 ln
$$\alpha^{\circ}_{kao-w} = 2.5 \cdot 10^6 \text{ T}^{-2} (\text{K}) - 2.87$$
 (3)

The equation for hydrogen isotopic fractionation between kaolinite and water was calculated by least squares regression using data from Sheppard *et al.* (1969) and Lambert and Epstein (1980), with a correlation coefficient of 0.999 and a standard error of 1.052 for $T \le$ 180°C:

$$1000 \ln \alpha^{H}_{kao-w} = 0.18 \text{ T (°C)} - 33.13$$
 (4)

where the subscripts w and kao and the superscripts O and H stand for water, kaolinite, oxygen, and hydrogen, respectively.

RESULTS

The results of the δ^{18} O and δ D analyses of kaolinite are presented in Table 1 and Figures 3-4. Stable isotope data for authigenic kaolinite and weathering kaolinite from Permian sandstones in the nearby Denison Trough and for kaolinite of pre-Middle Tertiary and Middle to Late Tertiary from elsewhere in eastern Australia (Bird and Chivas, 1988; Baker and Golding, 1992) are



Figure 3. δ^{18} O and δ D values of kaolinite with sample positions relative to the surface at Black Ridge. The numerals in the figure are δ^{18} O^{∞} values of the samples.

also shown in Figure 4 for comparison. Several characteristics may be recognized from the kaolinite stable isotopic data from the "White Section":

- low δ¹⁸O values with relatively small variation (12.3‰ to 14.8‰);
- very low δD values with a large variation (-120‰ to -85‰);
- 3) in Figure 4, two of the samples (304 and k2) are on the "kaolinite line," but one sample (309) is above the "supergene/hypogene" line, which clearly separates distributions of stable isotope data between shallow supergene and deep hypogene kaolinites according to a stable isotope study of kaolinite from the Santa Rita Deposit (Taylor, 1974, 1979);
- from the kaolinite line, the large variation in δD values appears to be negatively related to that of the δ¹⁸O values; and
- 5) the δD values have a positive correlation with depth from the present surface (Figure 3).

INTERPRETATION OF THE DATA

Age and origin of the weathering

Very low δ^{18} O and δ D values of kaolinite from eastern Australia have been reported by recent studies (Bird and Chivas, 1988, 1989; Baker and Golding, 1992). These studies demonstrate that the low isotope values are the result of isotopic equilibrium between kaolinite and meteoric water during the Late Paleozoic to Early Mesozoic, when Australia was at a higher latitude and the climate was more humid and cool. One of the characteristics of the kaolinite isotope data from the



Figure 4. Stable isotopic characteristics of kaolinites from the "White Section," Black Ridge, and from Permian sandstones in the Denison Trough. Filled circles = from Black Ridge. Open circles = from the weathered Permian sandstone, Springsure, Denison Trough (Bird = and Chivas, 1989). Note the "transported" kaolinites are not included. Filled downward triangles = authigenic kaolinite from the Aldebaran Sandstone (Baker and Golding, 1992). Open downward triangles = kaolinite from the weathered Aldebaran Sandstone (Baker and Golding, 1992). Squares = kaolinite of pre-Middle Tertiary regolith (Bird and Chivas, 1989). Upward triangles = kaolinite of Middle to Late Tertiary regolith (Bird and Chivas, 1989). A, B, C = the Late Triassic to Early Jurassic, pre-Middle Tertiary, Middle to Late Tertiary age groups, respectively; a, b, c = the corresponding meteoric water. Note the one sample of pre-Middle Tertiary age (square) in A group has some contamination of quartz (Bird and Chivas, 1989). d (dotted ellipses) = modern meteoric water in eastern Australia (from Bird and Chivas, 1989). Dotted line = the temperature fractionation line. ML = the meteoric water line (Craig, 1961). S/H = the supergene/hypogene line (Sheppard et al., 1969). KL = the kaolinite line (Savin and Epstein, 1970). Note the temperature fractionation line starting from sample 304 at 15°C intersects the authigenic kaolinite of the Aldebaran Sandstone at about 50°C.

"White Section" is that samples 304 and k2 plot on the "kaolinite line." For these samples, the δD values are -111% and -120%, which are the most negative values for kaolinites yet reported from weathering profiles in eastern Australia (Bird and Chivas, 1988, 1989). We suggest the above values, therefore, are isotopically unshifted weathering products that were in equilibrium with the ancient surface meteoric water. Australian regolith profiles have been investigated in detail using oxygen isotope data from clay minerals developed in these profiles (Bird and Chivas, 1988, 1989). According to their age category (Bird and Chivas, 1988, 1989), clay $\delta^{18}O$ values of less than 14–15‰ are thought to reflect weathering events of Early to mid-Mesozoic age. As shown in Figure 4, all kaolinites from the "White Section" and from the Permian sandstones in the Denison Trough have δ^{18} O values consistent with Early to mid-Mesozoic kaolinization and much lower than those of the "pre-Middle Tertiary" and "Middle to Late Tertiary" kaolinite from eastern Australia (see Figure 4).

A stable isotope study of authigenic kaolinite in equilibrium with meteoric water from the early Permian Aldebaran Sandstone in the Denison Trough gives $\delta^{18}O$ values from +7.8\% to 8.9\% between 45°C and 70°C (Baker and Golding, 1992). The δ^{18} O values of the corresponding meteoric water should be between -13.9% and -10.1%, averaging -12%. Baker and Golding (1992) concluded that deep penetration of this water into the Denison Trough is linked to Late Triassic structural inversion, which led to the exposure of the northern part of the rock unit by the Early Jurassic. The surface meteoric water should have been about 12-15°C during that time since central Queensland was at about 50-55°S during the Late Triassic to Early Jurassic (Smith et al., 1981; Veevers, 1984). At this temperature range, the δ^{18} O values of meteoric water in equilibrium with the kaolinite of samples 304 and k1 would have been -13.1% to -12.5% and -13.4% to -12.8% (see Table 1), respectively, averaging -13%calculated using Eq. 3. The calculated meteoric water value is almost identical to that calculated from Baker and Golding's data and implies that kaolinite in the "White Section" formed during the Late Triassic to Early Jurassic, or perhaps even earlier.

Variation trends in the $\delta^{18}O$ and δD values

The variation in δ^{18} O values of kaolinite from the "White Section" is 2.5‰, which may not be significant. The variation in δD values, however, is 35%, which is much larger than those of previous studies in eastern Australia (Bird and Chivas, 1988, 1989; Baker and Golding, 1992) and is too large for the system to be in isotopic equilibrium, considering the samples were collected from a restricted area (all within 250 m of each other). Comparing the stable isotopic data of the "White Section" kaolinite with those from Permian sandstones of the nearby Denison Trough and from elsewhere in eastern Australia (Figure 4), the following characteristics may be recognized: 1) for each age category, the O-isotopic variations are very small (for the one exception, see the notes in Figure 4), the H-isotopic variations are relatively large, and the stable isotopic shift from the kaolinite line is essentially vertical; 2) the H-isotopic shift becomes smaller in younger age category and, only in the Triassic to Early Jurassic group, some data are above the S/H line; 3) for such a small area, the "White Section" kaolinite has greater variation in δD and $\delta^{18}O$ values than in any age category from the regolith in eastern Australia, and the trend of the isotopic shift appears to be slightly negative rather than vertical in Figure 4.

One of the possibilities for the large δD variation is contamination of the samples by weathering products of different ages. This cannot explain the variation trends of δD values in Figures 3 and 4 because both the $\delta^{18}O$ and δD values of surface weathering kaolinite in eastern Australia should have become progressively higher since the late Paleozoic as a result of an increase in the δD and $\delta^{18}O$ values of meteoric waters when continental drift moved Australia from high to low latitudes, resulting in a progressively warmer climate (Bird and Chivas, 1988). Hence, both the trend slopes of the $\delta^{18}O$ and δD values should be positive in Figures 3 and 4 if they had formed by weathering during different periods.

Another possibility for the variation is a change of temperature either due to cooling of an ascending hydrothermal fluid or perhaps an abnormal temperature gradient with depth. The temperature change may cause variations in the same direction as the trend in Figures 3 and 4. The *pure* effect of temperature change, however, will not produce the same degree of variation as that shown by the samples in Figure 4. Moreover, assuming sample 304 was formed at surface conditions (i.e., about 15°C, see last section), for the same water to equilibrate kaolinite with a δD value of -85% such as sample 309, the temperature must have been about 160°C. From sample 304 to 309, the depth change is only 10 m, and it requires an unlikely thermal gradient to produce a change of 145°C temperature in 10 m, especially as there are no structural or petrological variations to indicate such an abnormal gradient.

Variations of the δ^{18} O and δ D values of kaolinite in the "White Section" may be interpreted as due to mixing of a kaolinite of weathering origin (samples 304 and k2) with different proportions of diagenetic kaolinite with isotopic values of $\delta^{18}O < 12.3\%$ and δD > -85% (samples 207, 309 and k1). Microscopic observation indicates that there appear to be two generations of kaolinite in the "White Section" (Figure 2). The proposed earlier generation of kaolinite, however, is too small in volume to cause the variation in the observed δD values. Nevertheless, it is impossible to verify different kaolinite origins microscopically, so this assumption cannot be totally ruled out, even though it needs relatively large proportions of diagenetic kaolinite with very heavy δD values and light $\delta^{18}O$ values in the "White Section."

Longstaffe and Avner (1990) have suggested that H-isotope exchange can occur independently of O-isotope exchange in sandstones at temperatures as low as 40°C. Therefore, the large variation in the δD values and the isotope variation trends of the "White Section" kaolinite in Figure 3 and 4 may be due to post-formation, partial isotopic exchange. The partial isotopic exchange in δD of the kaolinite could be affected by later heavier meteoric water. The δD value of the meteoric water corresponding to the pre-Middle Tertiary weathering should be about -62% (Figure 4). At 40°C, even if the kaolinite had been totally reequilibrated with a large amount of meteoric water, it should be about -88%, i.e., still lower than that of sample 309. At 80°C, the kaolinite needs more than 85% exchange with the meteoric water to derive the δD value of sample 309. For the Middle to Late Tertiary meteoric water, the δD value should be about -43%; the kaolinite needs about 60% and 53% exchange with the meteoric water to produce the δD value of sample 309 at 40°C and 80°C, respectively. From this discussion, the later case may explain the large variation in δD values although this model needs a large amount of meteoric water and it still cannot account for the trends in Figures 3 and 4.

Another possibility is that the partial isotopic exchange was caused by reaction of kaolinite with fluids other than meteoric water. A recent geochemical study (Zhou, 1992) indicates that there was hydrothermal activity in the Clermont area after the Permian sedimentation. The fluid temperature was between 100°C to 150°C at the unconformity between the Anakie metamorphics and the conglomerates and was presumably much lower than 100°C in the overlying "White Section," which is about 80 m above the unconformity. This temperature is not high enough to affect the $\delta^{18}O$ values significantly, but sufficient for exchange of H-isotopes in kaolinites. Stable isotopic studies suggest that the heated fluid in the conglomerates was dominated by marine water (Zhou, 1992). In this case, 30% exchange with this water would produce the δD value of sample 309 at 40°C. The upward movement of this fluid should be affected by the permeability of the "White Section." Petrological and mineralogical studies indicate that the alteration only very slightly affected the lower part of the "White Section" (Zhou, 1992), and it may have never reached the upper parts of the "White Section," i.e., the positions of samples 304 and k1. Hence, the partial exchange of hydrogen isotopes between this fluid and kaolinites in the lower parts of the "White Section" may cause a large variation in δD values and only slight variation in $\delta^{18}O$ values, and the trends in Figures 3 and 4 may reflect different proportions of this fluid which reacted with the "White Section."

DISCUSSION

Although there are several possible ways to interpret the results of the kaolinite isotopic analyses, it is most likely that the large variation in δD values are the result of post-formational isotopic exchange of fluid with kaolinites of weathering origin. Mixing with diagenetic kaolinite may also have contributed, to some extent, to the observed variation. In any case, kaolinites from samples 304 and k2 are on the supergene kaolinite line, and their δD and $\delta^{18}O$ values are generally consistent with an origin as the original weathering products.

Sample no.	Latitude	Longitude	δ ¹⁸ O‰	δD‰
NQLD-6 ¹	24°2′S	148°3′	12.5	
NQLD-8A ¹	24°2'S	148°2'	13.5	-80
NOLD-8A ¹	24°2′S	148°2'	14.3	-81
SS10-32	24°3′S	148°1′	14.6	-93
SS4-5 ²	24°6′S	148°4'	11.7	-104
ED4-17 ²	25°0′S	148°3′	11.8	

Table 2. Stable isotopic data for kaolinite from weathered Permian sandstone, Springsure, Denison Trough.

¹ Data from Bird and Chivas, 1989.

² Data from Baker and Golding, 1992.

Compared with the kaolinite stable isotopic data from the Denison Trough and the eastern Australian regolith (Figure 4), samples 304 and k2 appear to represent a surface weathering product during the Late Triassic to Early Jurassic. The "White Section," therefore, should have formed pre-Early Jurassic, most likely pre-Late Triassic, which totally dismisses the Tertiary age of the rock unit itself (Osman and Wilson, 1975) and the assumption of Permian diagenetic or Tertiary leaching origin for the kaolinite alteration (Beeston, 1978). Considering a previous sedimentary study (Cook and Taylor, 1979), a late Permian age for the "White Section" may be most appropriate.

The large variation in δD values of the "White Section" kaolinite may be interpreted as due to post-formational isotopic exchange with later meteoric water and/or the heated fluid from the underlying conglomerates. However, the trends of the isotopic shift in Figures 3 and 4 appear to favor the later. Nevertheless, since the trends are derived from only five samples, an unrevealed vertical shift would be possible if the database were much larger. Therefore, a later meteoric model cannot be totally dismissed.

It is notable that kaolinite δ^{18} O values in the "White Section" are virtually identical to those of weathered Permian sandstone in the Denison Trough (Table 2); the slightly lower δ^{18} O values of some of Baker and Golding's (1992) samples may due to contamination of authigenic kaolinite (Baker, personal communication, 1992). The much higher δD values and restricted variation of kaolinite data from the Denison Trough may be due to post-formational equilibrium δD exchange. The authigenic kaolinite in the early Permian Aldebaran Sandstone in the Denison Trough was formed in equilibrium with meteoric water, which displays virtually the same stable isotopic values as that producing the weathering kaolinite discussed above, i.e. they were all formed about the same time. Since the Permian basins in the Clermont gold field are tectonically aligned with the Denison Trough (Dickins and Malone, 1973) and the stable isotopic characteristics of kaolinites from the "White Section" and the Denison Trough are comparable, it is quite possible that these small basins in the Clermont area were part of the Denison Trough. Preservation of the original weathering δD values of samples 304 and k2 implies that the Miclere-Black Ridge basin was not deeply buried after the Late Triassic inversion. This contrasts with the largely shifted δD values of kaolinite in the Permian sandstone in the Denison Trough (Table 2), which was reburied by more than 1000 m of Surat Basin sediments during the Jurassic and Early Cretaceous. These characteristics may partly explain the tectonic link between the Permian Miclere-Black Ridge basin in the Clermont area and the Denison Trough and the fact that no Mesozoic sediments are observed in the Miclere-Black Ridge basin and other small Permian basins in the Clermont area (Dickins and Malone, 1973).

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