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Origin of sedimentary kaolin in the Neuquén basin, Argentina as determined by oxygen isotopes

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ABSTRACT. — Two Gondwana granite-rhyolite provinces occur in southern Argentina. These are, Choiyoi, late Carboniferous-Early Permian to Triassic in age, in the Neuquén basin and Chon-Aike, Middle Jurassic, in Patagonia. In both areas, the clay facies of a fluvial sequence that overlay the complexes is composed of transported kaolinites. In Patagonia, a paleoweathered surface was formed on Chon-Aike rocks under humid temperate conditions indicated by the oxygen isotope composition of the kaolinites ($\delta^{18}O$ =16-18‰). In this work it is demonstrated that the isotopic composition of the kaolinites eroded from that paleosurface and deposited in a fluvial sequence has not changed. Taking this into consideration, the $\delta^{18}O$ values obtained in the sedimentary kaolinites of the Neuquén basin (15 to 17‰), can be considered as an indication of humid, temperate conditions as the primary origin of the kaolinite. The clasts composing the coarser facies (rhyolitic) indicate that the kaolinite has been eroded from a paleoweathered surface developed on Choiyoi rocks.

This work demonstrates that following a huge volcanic event in the southern hemisphere, paleoweathered surfaces developed in a humid and temperate climate, conditions already established for Chon-Aike kaolinites. Moreover all the continental sedimentary sequences that overlay Choiyoi and Chon-Aike are promising areas for prospecting for sedimentary kaolin deposits.

RIASSUNTO. — Nell'Argentina meridionale sono presenti due province della Gondwana caratterizzate dalla presenza di associazioni granitico-riolitiche: quella di Choiyoi, di età tardo Carbonifero-Triassica, nel bacino del Neuquén e quella medio Giurassica di Chon-Aike, in Patagonia. La facies argillosa della sequenza fluviale che ricopre i prodotti magmatici di entrambe le aree è composta di caoliniti rimaneggiate. La composizione isotopica dell'ossigeno delle caoliniti ($\delta^{18}O$ = 16-18‰) suggerisce che la paleosuperficie di alterazione delle rocce di Chon-Aike è da attribuire ad ambienti temperati-umidi. I risultati del lavoro evidenziano che durante i processi di erosione della paleosuperficie e sedimentazione in ambiente fluviale, la composizione isotopica dell'ossigeno non ha subìto cambiamenti. Su queste basi, i valori di $\delta^{18}O$ delle caoliniti sedimentarie del bacino di Neuqén (15-17‰) possono essere considerati indicatori di ambienti analoghi a quelli delle rocce di Chon-Aike. I clasti che compongono la facies più grossolana (riolitica) indicano che la caolinite deriva dall'erosione di una paleosuperficie di alterazione sviluppatasi a spese delle rocce di Choiyoi. Questo lavoro dimostra che le paleosuperfici di alterazione si sono sviluppate in ambienti climatici temperati-umidi, come già ipotizzato per le caoliniti di Chon-Aike. Tutte le sequenze sedimentarie continentali

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che ricoprono le aree di Choiyoi e Chon-Aike rappresentano possibili siti per lo sfruttamento di caolino sedimentario.

KEY WORDS: *Oxygen isotopes, kaolinite origin.*

INTRODUCTION

Two Gondwana granite-rhyolite provinces occur in southern Argentina; Choiyoi, late Carboniferous-Early Permian to Triassic in age, in the Neuquén basin and Chon-Aike, Middle Jurassic, in Patagonia (fig. 1B). In both areas, the clay facies of a fluvial sequence that overlays the complexes is composed of transported kaolinites.

In Patagonia, a paleoweathered surface was formed on Chon-Aike rocks under humid - temperate conditions as indicated by the oxygen isotope composition of kaolinites (16-18), Cravero *et al.*, 1991, Cravero and Domínguez 1992a. The material from this surface (mainly kaolinitic) was transported by streams and deposited during the lower Cretaceous in what is known as the Baqueró Formation (Cravero and Domínguez, 1992b). Cravero and Domínguez (1991) demonstrated that during transportation, kaolinite crystals were broken producing a decrease in the grain size and crystallinity. These deposits are ideal to determine if the oxygen-isotope values are modified when the kaolinite has experienced changes during its transportation.

In the Neuquén basin, the Middle Jurassic fluvial sequence, where the clay facies is composed of transported kaolinites, is termed the Challacó Formation. The source rocks where the primary alteration took place have not yet been identified, nor has the alteration process (hydrothermal or weathering) that led to the kaolinite formation.

In both areas the clay facies are kaolin deposits that are mined for ceramic purposes. In any area where kaolin deposits are exploited, the source rocks and their type of alteration strongly influences the development of mining operations.

Although several methods have been proposed to distinguish between meteoric and hydrothermal kaolinite, oxygen isotopes appear to be the most powerful tool for that discrimination.

The goal of this study is to determine the source rocks and the primary process that produced the kaolinites that form the clay facies of the Challacó Formation in the Neuquén Basin. $\delta^{18}O$ of these kaolinites was used to determine the origin of the primary alteration in the source area after showing that the isotopic composition of the kaolinites was unchanged during transportation. To prove this, $\delta^{18}O$ in kaolinites from the Baqueró Formation was determined to compare with values already known from the Choin-Aike paleoweathered surface. The coarser facies of the Challacó Formation, sandstones and conglomerates were used to determine the type of rocks from which the kaolinization took place. Domínguez, 1988 suggested that kaolinite could have originated from an extended weathered area developed on rhyolitic volcanic rocks Triassic in age (Choiyoi rocks) although some hydrothermal kaolinite manifestations are well known in these rocks (Domínguez 1990, Zolner and Amos 1973). If the existence of a large weathered surface is proved, then clues for further exploration in the area would be provided.

Evidences provided from this study show that the kaolin oxygen isotopic composition does not change during transportation and deposition and also that the Neuquén kaolin deposits were formed by the erosion of a large weathered area formed in warm climatic conditions in the lower Jurassic.

GEOLOGICAL BACKGROUND

In the Neuquén Basin, the kaolin deposits that are the clay facies of the Challacó Formation are distributed in an area of about 1000 km^2 (fig. 1A). Clay production began in 1940 and in recent years the annual average output has been over 40.000 mT and the kaolin resources are over 30 mmT.

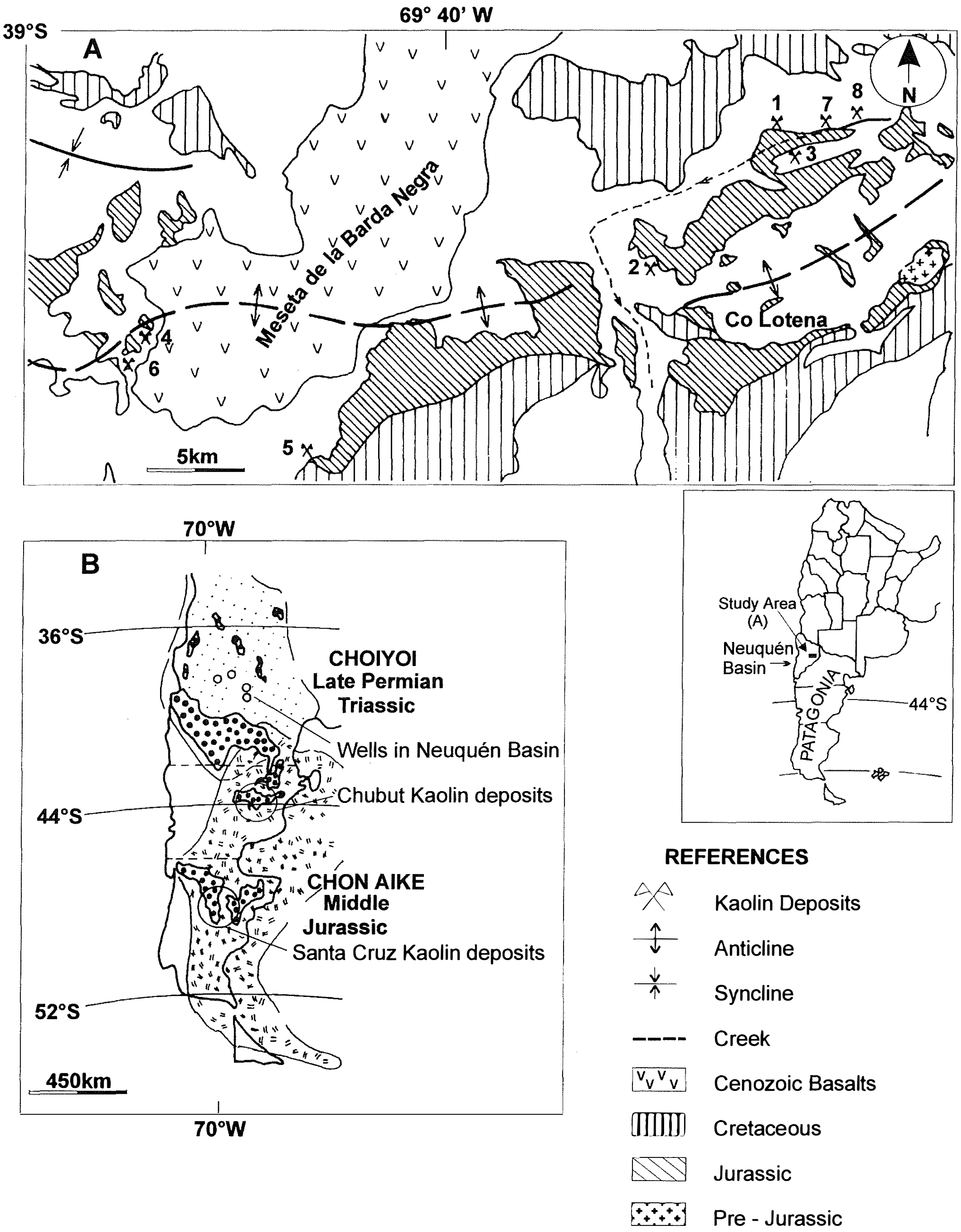

Fig. 1A – Geological setting of the kaolin deposits in the study area of Neuquén Basin. After Zavala, 1993. Sampling localities are marked as 1, 2 etc.; B – Distribution of Chon-Aike and Choiyoi Provinces in Patagonia after Key *et al.*, 1989. Outcrops are indicated as symbols enclosed in full lines.

The basement of this basin is a granite-metamorphic complex covered and intruded by late Corboniferous-Early Permian to Triassic granites and rhyolites termed the Choiyoi province. This complex coupled with the Upper Triassic Granites to Middle Jurassic rhyolites of Patagonia, termed Chon-Aike are two of the most prominent Gondwana granite-rhyolite provinces in South America (Key *et al.*, 1989, figure 1B). The Neuquén basin began to develop during the Jurassic Extension. The Jurassic interval of the basin is represented by three sedimentary cycles (Precuyano, Cuyano and Loteniano-Chacayano). The Cuyano cycle represents the first marine deposition and is composed of three formations: Los Molles (deep-sea facies), Lajas (shallow and platform facies) and Challacó (fluvial facies) (Gulisano *et al.*, 1984). This sequence unconformably rests either on Choiyoi rocks or on other minor pyroclastic deposits such as Chacaico Formation (Hettangian-Sinemurian).

The Challacó Formation crops out in a wide area where kaolin mines have been developed. The clay facies are massive and varied in color, red, yellow, brown and gray to white. The thickness ranges from 4 to 16 m and are laterally constant so that the kaolin can be followed for hundred of meters. The clay bearing formation strikes 75-100° and dips 8-10°N. The coarser facies of the formation include sandstones and conglomerates. In the area of the deposits, the Challacó Formation corresponds to an anastomosed fluvial system developed during late Bajocian-Middle Bathonian, Middle Jurassic (Zavala, 1993). This formation is unconformably covered by flat lying Cretaceous sediments and Cenozoic basalts.

In southern Argentina, Patagonia, the sedimentary kaolin deposits of the Baqueró Formation, lower Cretaceous in age, crop out mainly in the Santa Cruz kaolin deposits area (fig. 2B). This Formation overlays the Chon-Aike rocks, Middle Jurassic in age.

Chon-Aike is composed of rhyolitic ash-flow tuffs. These rocks crop out in a wide area in southern Argentina (Patagonia) where Cravero *et al.* (1991) have indicated the existence of an Upper Jurassic-Lower Cretaceous paleoweathered surface with residual kaolin deposits (Santa Cruz and Chubut kaolin deposits area, fig. 1B). The Baqueró Formation is composed of fining-upward sequences where the clay facies are at the top. The conglomerates and sandstones are composed of volcanic-pyroclastic clasts from Chon-Aike rocks with a kaolinitic clay matrix. The 5-7 mts thick grey to brownish clay facies are lenticular and constitute the kaolin deposits now being mined. The kaolinite was eroded and transported from the Chon-Aike paleoweathered surface by meandering streams (Cravero and Domínguez, 1992b).

MATERIALS AND METHODS

Clay point samples from different outcrops, mainly mines, in the Challacó Formation were analyzed. Figure 1A shows the sampling locations. Three samples were also studied from the Baqueró Formation outcropping in the Santa Cruz kaolin deposits area (fig. 1B), where the kaolinite cristallinity was modified during transportation.

The mineralogical composition of the clay fraction was determined by XRD using glass slides on which suspensions were obtained by standard sedimentation techniques. The I/S type was determined after glycolation with ethylen-glycol using the method described in Moore and Reynolds, (1989). The mineralogy of the >44 μm fraction was determined microscopically.

For the isotopic determination the <2 μm clay fraction was separated by standard sedimentation techniques. No dispersant was added in order to avoid interferences during isotopic analysis. Where necessary, the Fe-oxides and -hydroxides were removed using the method of Mehera and Jackson (1960). The purity of the samples was checked using XRD. In those where mixed layer I/S was found, the <0.2 μm fraction was extracted because the I/S is concentrated in the finest fractions. The

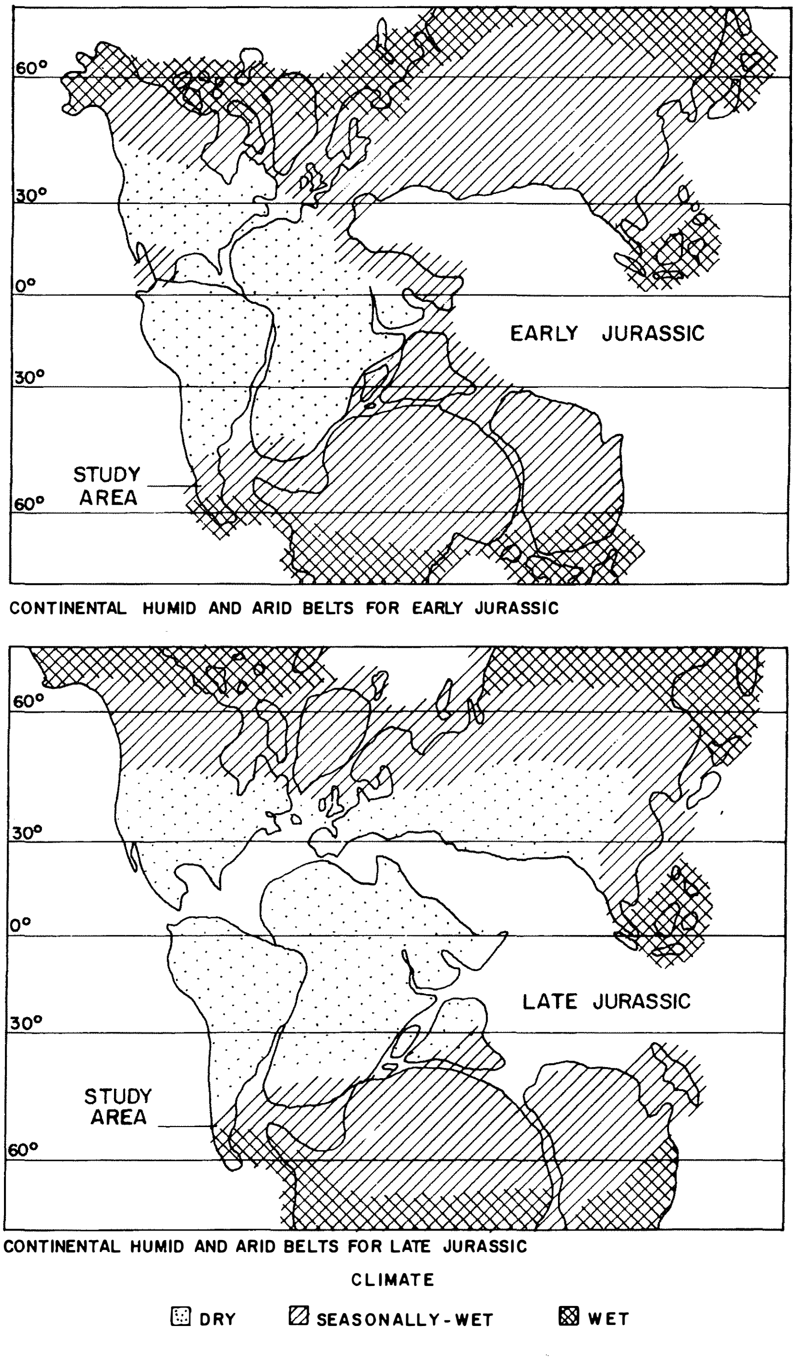

Fig. 2 – Continental humid and arid belts for Jurassic (After Hallam, 1985).

semi-quantitative mineralogical composition was calculated following the Chung (1974) method (Table 1).

The oxygen isotopic composition was determined in both the 2-0.2 μm and less than 0.2 μm fraction in order to determine the interference that I/S might introduce in the $\delta^{18}O$ values.

Oxygen, for isotopic analysis was liberated quantitatively from dried and thoroughly outgassed 10 mg samples by reaction with bromine pentafluoride at 550° (Clayton and Mayeda, 1963). Oxygen thus liberated was quantitatively converted to carbon dioxide by Pt-Catalyzed reaction with an incandescent carbon rod and the $\delta^{18}O$ ratio of the resultant gas measured using a modified MS-12 dualinlet triple-collecting mass spectrometer. Precision of $\delta^{18}O$ measurements is ±0.1%. and the values are reported relative to Vienna Standard Mean Ocean Water (V-SMOW). The standard used was MBS 28 and ANU Si.

RESULTS

In the Challacó Formation the clays are composed of 50% to 98% kaolinite, 2 to 5% quartz, and 50% to 0 mixed I/S layers. In these interestratified clays about 80% corresponds to Illite. The amount of I/S increases in the fraction less than 0.2 μm. The coarser fractions of the clay facies (>44 μm) are composed of quartz (volcanic shape), feldspar, plagioclase

TABLE 1

$\delta^{18}O$ data and mineralogical composition of sedimentary kaolinite samples from Challacó and Baqueró Formation. Formation. Sampling locality (1, 2, etc.) in Neuquén deposits is shown in fig. 1A. kaol.: kaolinite, I/S: mixed layer illite-smectite, Qtz: quartz.

Locality	Deposit	Formation	Area	Grain saize(μm)	$\delta^{18}O$	Kaol. %	I/S %	Qtz %
1	Las Mellizas	Challacó	Neuquén	2-0.2	16.4	82.0	16.0	2.0
1	Las Mellizas	Challacó	Neuquén	2-0.2	16.2	82.0	16.0	2.0
1	Las Mellizas	Challacó	Neuquén	<0.2	16.5	55.0	45.0	-
2	El 25	Challacó	Neuquén	<0.2	17.9	54.0	46.0	-
3	La Sociedad	Challacó	Neuquén	2-0.2	18.4	79.0	19.0	2.0
3	La Sociedad	Challacó	Neuquén	< 2	17.7	68.0	32.0	-
4	Tatacha	Challacó	Neuquén	2-0.2	16.1	73.0	24.0	3.0
4	Tatacha	Challacó	Neuquén	<0.2	17.5	57.0	43.0	-
5	Mercedes	Challacó	Neuquén	2-0.2	15.6	62.0	36.0	2.0
6	Verdacho	Challacó	Neuquén	2-0.2	17.0	48.0	47.0	5.0
7	La cuña	Challacó	Neuquén	2-0.2	17.5	80.0	14.0	6.0
8	La Beatriz II	Challacó	Neuquén	<2	18.8	95.0	-	5.0
-	Camp 2	Baqueró	Santa Cruz	<2	16.3	96.0	-	4.0
-	Camp 1	Baqueró	Santa Cruz	<2	16.2	95.0	-	5.0
-	Camp 1	Baqueró	Santa Cruz	<2	17.1	96.0	-	4.0

TABLE 2

Oxygen-isotope composition of kaolinites coming from the alteration of Chon-Aike rocks in Chubut and Santa Cruz areas and of kaolinites transported and deposited in Baqueró Formation.

Formation/Age	Deposit	Area	Origin	$\delta^{18}O$
Chon-Aike/Middle Jurassic	Lote 8	Santa Cruz	Residual	18.2
Chon-Aike/Middle Jurassic	Lote 8	Santa Cruz	Residual	17.4
Chon-Aike/Middle Jurassic	Lote 8	Santa Cruz	Residual	16.8
Chon-Aike/Middle Jurassic	Lote 18	Santa Cruz	Residual	17.5
Baqueró/Lower Cretaceous	Camp 2	Santa Cruz	Sedimentary/Transported	16.4
Baqueró/Lower Cretaceous	Camp 1	Santa Cruz	Sedimentary/Transported	16.2
Baqueró/Lower Cretaceous	Camp 1	Santa Cruz	Sedimentary/Transported	17.1
Chon-Aike/Middle Jurassic	Romina	Chubut	Residual	18.7
Chon-Aike/Middle Jurassic	C-60	Chubut	Residual	18.3
Chon-Aike/Middle Jurassic	Valeriana	Chubut	Residual	17.2
Chon-Aike/Middle Jurassic	D.Carlos	Chubut	Residual	16.1

and variable amounts of iron oxides including magnetite and hematite. Volcanic glass and pyrite nodules are also found in some deposits. Conglomerate clasts are mainly composed of rhyolitic rocks and granites in smaller amounts. The clay fraction mineralogy in the Baqueró Formation samples (<2 μm) is composed of about 95% kaolinite and 5% quartz. The coarser grains (>325 μm) are composed of quartz and minor amounts of feldspars and lithic clasts

The isotopic values obtained in kaolinites of the Challacó and Baqueró Formations are shown in Table 1. Table 2 shows the values obtained for the transported kaolinites of the Baqueró Formation plus the data from Cravero *et al.*, (1991) in the residual kaolinites of the Chon-Aike rocks from the Santa Cruz and Chubut kaolin deposits (fig. 1B). The Chon-Aike primary kaolinites and the transported ones of the Baqueró Formation show similar values, ranging from 16 to 18. The samples from Challacó Formation show a very similar isotopic composition, not only between different samples but also between different fractions of the same sample.

DISCUSSION

Savin and Epstein (1970) demonstrated that kaolinite forms in isotopic equilibrium with its environment, therefore, if no post-formation changes occur, the isotopic composition of the kaolinite would reflect the conditions existing at the time of kaolinite formation.

Most of the weathering products in the continental USA have values of $\delta^{18}O$ of about 15-25. This general range is accepted for weathering elsewhere because the research covered the total spectrum of climatic conditions. In addition, because the isotopic composition of the oceans has been relatively unchanged over geologic time, this range also applies to the past (Lawrence and Taylor, 1971). For ancient clays, only oxygen-isotope datum is useful, because as Bird and Chivas (1988) and O'neill and Kharaka (1976) have shown, hydrogen is more sensitive than oxygen to isotopic exchange with ground water when the clays are very old. Bird and Chivas (1993) used the oxygen isotope composition of kaolinite to establish climatic changes as Australia drifted northwestward during its geological history.

The isotopic composition of the sedimentary Baqueró Formation kaolinite (Lower Cretaceous) is similar to that determined for the residual kaolinite formed in a nearby weathered area (Middle to Lower Jurassic). Although in these sedimentary kaolinites a lowering in their grain size and crystallinity during transportation was found (Cravero and Domínguez, 1991), the isotopic composition is unchanged (Table 2). This conclusion allows the use of the oxygen isotopic composition of transported kaolinites to ascertain the processes that formed the primary ones.

In the Challacó Formation the clay facies were deposited on an alluvial plain. The mineralogical and petrographical composition of the sandstone and conglomerate facies of the fluvial sequence, mainly rhyolites, indicate that the source area of these rocks was the granite-rhyolite complex of Choiyoi. (Upper Carboniferous - Upper Triassic) that covers a wide area of the Basin (fig. 1B) because no other older lithological unit bears this type of rocks.

The homogeneity in the isotopic values agrees with a unique source for the kaolinite. Cravero *et al.*, (1997) determined that the I/S was formed in the depositional basin by the transformation of illite which was eroded from the source area. The isotopic values of the kaolinite (Table 1) appear not to be influenced by the presence of I/S, so the isotopic composition of the I/S must be very close to that of the kaolinite. This permits to consider that the isotopic composition of the whole sample is indicative of the isotopic composition of the kaolinite. Because transportation does not change the original isotopic composition of kaolinite the obtained values of $\delta^{18}O$ of about 16-18 must indicate the conditions under which the kaolinite was formed. The kaolinite could have come either from the meteoric alteration of the Choiyoi rocks or from the hydrothermal alteration of such rocks. Widespread hydrothermal gold mineralization with kaolinized wall rocks is known in the Cordillera del Viento (about 200 km north of the study area, fig. 1B) where these rocks outcrop. Oxygen isotope composition of kaolinite from a hydrothermal gold deposit formed in Choiyoi shows low values, as expected, of 11 for $\delta^{18}O$ with a formation temperature of 210°C (Domínguez, 1990). The values obtained in this work are higher and clearly are related to meteoric waters. These values are also similar to those of Patagonia (Cravero *et al.*, 1991) probably indicating similar climatic conditions, humid and temperate. Therefore the development of a paleoweathered area is expected to have been developed on the Choiyoi rocks. The ages of the volcanic rocks and the sedimentary sequence indicate that the weathering process was developed in a time span between the Upper Triassic and the Middle Jurassic, probably the lower Jurassic. There are no known outcrops of this paleosurface.

Humid or at least seasonally humid conditions are well known for this part of the world during Jurassic times (Hallam, 1985, fig. 2). Warm to temperate uniform climatic conditions in those times for south America are reported also by Volkheimer, (1967).

The isotopic values in this area are quite similar to those found in the weathered area formed during Middle Jurassic-Lower Cretaceous times in the volcanic rocks of Chon-Aike in Patagonia, southern Argentina (Table 2). That means that during most of the Jurassic the climatic conditions were very similar in most of the southern part of Argentina. The difference in the age of the weathering, Lower to Middle Jurassic or Middle Jurassic to Lower Cretaceous depends mainly on the availability of source rocks for kaolinization and the geological history of the area. When the Neuquén basin began to develop (Early Jurassic), erosion from inland began and the kaolinite formed was eroded from the source by fluvial waters and deposited as overbank units. In Patagonia, Chon-Aike rocks were developed during Middle Jurassic to Late Jurassic and during the Lower Cretaceous, intracratonic basins were developed and fluvial waters eroded the weathered areas, depositing the kaolin clays mainly in ox-bow lakes of the meandering system (Baqueró Formation), (Cravero and Domínguez, 1992b)

CONCLUSIONS

This study shows that following a huge volcanic event in the Southern Hemisphere in Triassic-Jurassic times the subsequent climatic conditions were humid and temperate. Moreover all the continental sedimentary sequences that overlay Choiyoi and Chon-Aike are good areas for prospecting for sedimentary kaolin deposits in the Neuquén basin and Patagonia.

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