

A preliminary sulphur and oxygen isotope study of the Maha Sarakham evaporitic anhydrite from the Bamnet Narong Area of Northeastern Thailand.

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Abstract: The sulphur and oxygen isotopes of 10 anhydrite samples from 5 anhydrite beds of the Maha Sarakham Formation of Thailand (Basal Anhydrite, Basal Cap Anhydrite, Middle Anhydrite L, Middle Anhydrite U and Upper Anhydrite U) were investigated. This probably is the first time a Southeast Asian evaporite deposit has been studied. The isotopic compositions of both sulphur and oxygen of these anhydrites show a narrow variation (e.g., 14.5 to 16.5‰ with an average of 15.4‰ for the $\delta^{34}\text{S}$ value and 11.0 to 14.2‰ with an average of 12.2‰ for the $\delta^{18}\text{O}$ value) reflecting a fairly homogeneous source. There is no systematic change in both $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values with respect to the stratigraphic position. There was also no distinct difference in those values between the cap anhydrite and the normal anhydrite. The isotopic and geologic data suggest that the $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values of the anhydrite represent the original isotopic compositions of sulphate mineral precipitated from the restricted Khorat sea and also those in equilibrium with the world ocean sulphate during that geologic time period. Input and output of sulphur and oxygen through the sedimentary sulphur cycle, namely, reduction of seawater sulphate, crystallization of sulphate minerals, dissolution of pre-existing evaporites and oxidative weathering of old sulphides, were negligible in the restricted Khorat sea. Using the fairly well-established sulphur isotope age curve for the world sulphates is the late Mesozoic and Cenozoic and the average $\delta^{34}\text{S}$ value of the Maha Sarakham anhydrite together with the reported age of the underlying Khok Kruat Formation, the age of the Maha Sarakham evaporites in the Bamnet Narong area can best be estimated to be around Cenoman (lower part of Upper Cretaceous). The oxygen isotopic compositions of these anhydrites are slightly different from those of the best estimated values of world evaporites during this age. The new isotopic data should be added to the list for future revision of the curve because of the scarcity of previous data base in the construction of the curve.

INTRODUCTION

The evaporite deposits in the Khorat Plateau of the northeastern Thailand are well known as some of the world's major reserves of potash and rock salts. Many of the exploratory as well as detailed drilling programs in several areas in the Khorat Plateau were conducted by the Thai Department of Mineral Resources in the last decade to delimit areas of promising reserves. A number of studies on regional geology, subsurface geology of specific areas, mineralogy, geochemistry and petrography of selected drill cores were conducted to delineate the potash deposits and ultimately to understand the depositional environment and post-depositional changes affecting the evaporite deposits. This work has been carried out by many workers (Hite, 1974; Japakasetr, 1978; Japakasetr and Workman, 1978; Hite and Japakasetr, 1979; Japakasetr, 1980; Yumuang, 1983; and Hite, 1982).

This study was conducted independently with the primary aim of reporting, for the first time, the sulphur and oxygen isotopic compositions of the evaporites in this

part of the world, and of using the isotopic data to understand the geochemistry of the evaporite deposits as well as placing some constraints on their ages of formation.

GEOLOGY AND STRATIGRAPHY

The Khorat Plateau is a large blocklike platform covering an area of approximately 170,000 km² in northeast Thailand and central Laos (Fig. 1). The Plateau comprises two broad structural basins, the Sakhon Nakhon Basin in the north and the Khorat Basin in the south, whereas the Phu Phan uplift or anticlinorium which trends east-west across the Plateau, divides the north and the south Basins.

The potash and rock salt layers occur within the Maha Sarakham Formation. This Formation, which was mapped as the upper most rock unit of the Khorat Group (Japakasetr and Workman, 1981), is essentially marine evaporites intercalated with red clastics (Fig. 1). The underlying Formations of the Khorat Group, namely Khok Kruat, Phu Phan, Sao Khua, Phra Wihan, Phu Krudung and Nam Phong Formations, in descending order, are predominantly red-bed facies ranging from conglomerate (sandstone, siltstone) to shale with some intraclastic limestone.

The sedimentary sequences of the Maha Sarakham rock salt deposits in Bamnet Narong area can be classified into two-types I and II (Figs. 2 and 3). The type I sedimentary sequence is composed of three evaporitic cycles, the Basal Salt, Middle Salt and Upper Salt with three intercalated mudstone and claystone beds. A nearly complete evaporitic sequence ranging from iron oxides to K-Mg Salts is present in the Basal Salt cycle (Fig. 2; Yumuang, 1983). There is only one anhydrite bed, the Basal Anhydrite, in the Basal Salt cycle whereas two layers are observed in the Middle Salt cycle, the Middle Anhydrite L and the Middle Anhydrite U, and another two layers in the Upper Salt cycle, the Upper Anhydrite L and the Upper Anhydrite U, (Fig. 2).

The type II sedimentary sequence consists of only one Basal Salt cycle that contains two anhydrite layers, the Basal Anhydrite and the Basal Cap Anhydrite (Fig. 3). This type II sequence is commonly encountered in the drill holes near the crest of salt dome (Yumuang, 1983). Detailed petrographic description of each layer in these sedimentary sequences are given in Yumuang (1983) and Yumuang *et al.* (1985). It should, however, be noted that except for the Basal Cap Anhydrite bed, all the anhydrite layers show a nodular to finely laminated texture with calcareous algal and thin carbonaceous layers. The Basal Anhydrite bed is also relatively thin (0.77 to 1.86 meter thick) and rather persistent throughout the Khorat and Sakon Nakorn Basins (Hite and Japakasetr, 1979). It is, therefore, believed that these anhydrite layers are the origin depositional sequences (Yumuang *et al.*, 1985).

The Basal Cap Anhydrite, on the contrary, shows nodular mottled and gneissoid texture. The bed is tightly packed and strongly fractured with interbands of black carbonaceous layers (Yumuang, 1983). This cap anhydrite bed is believed to be a residual accumulation of anhydrite originally disseminated or interbedded within the upper part of the basal halite layer by the process of groundwater solutioning (Hite and Japakasetr, 1979; Hite, 1982; Yumuang *et al.*, 1986).

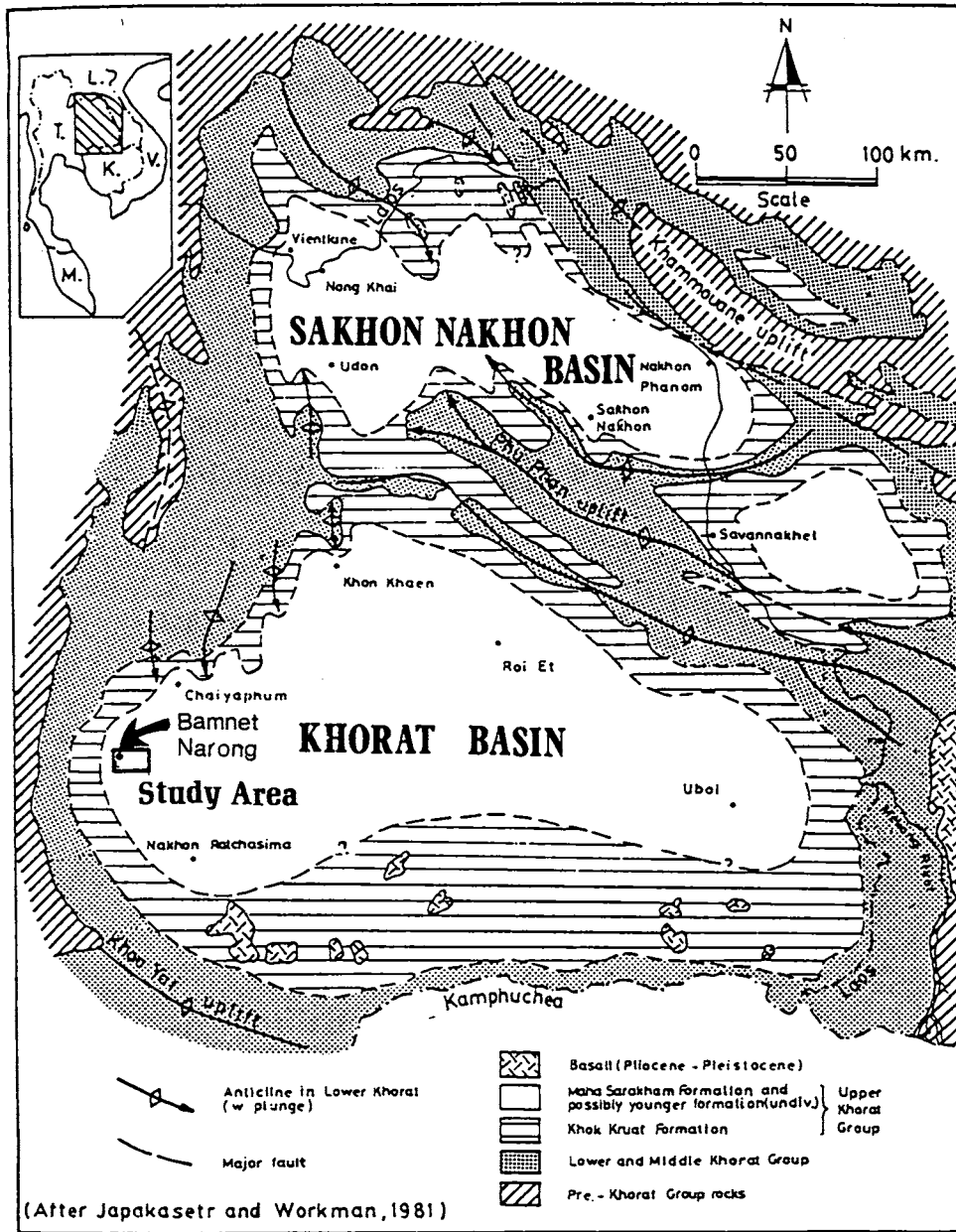


Fig. 1. Generalized geologic map of the northeast Thailand and central Laos.

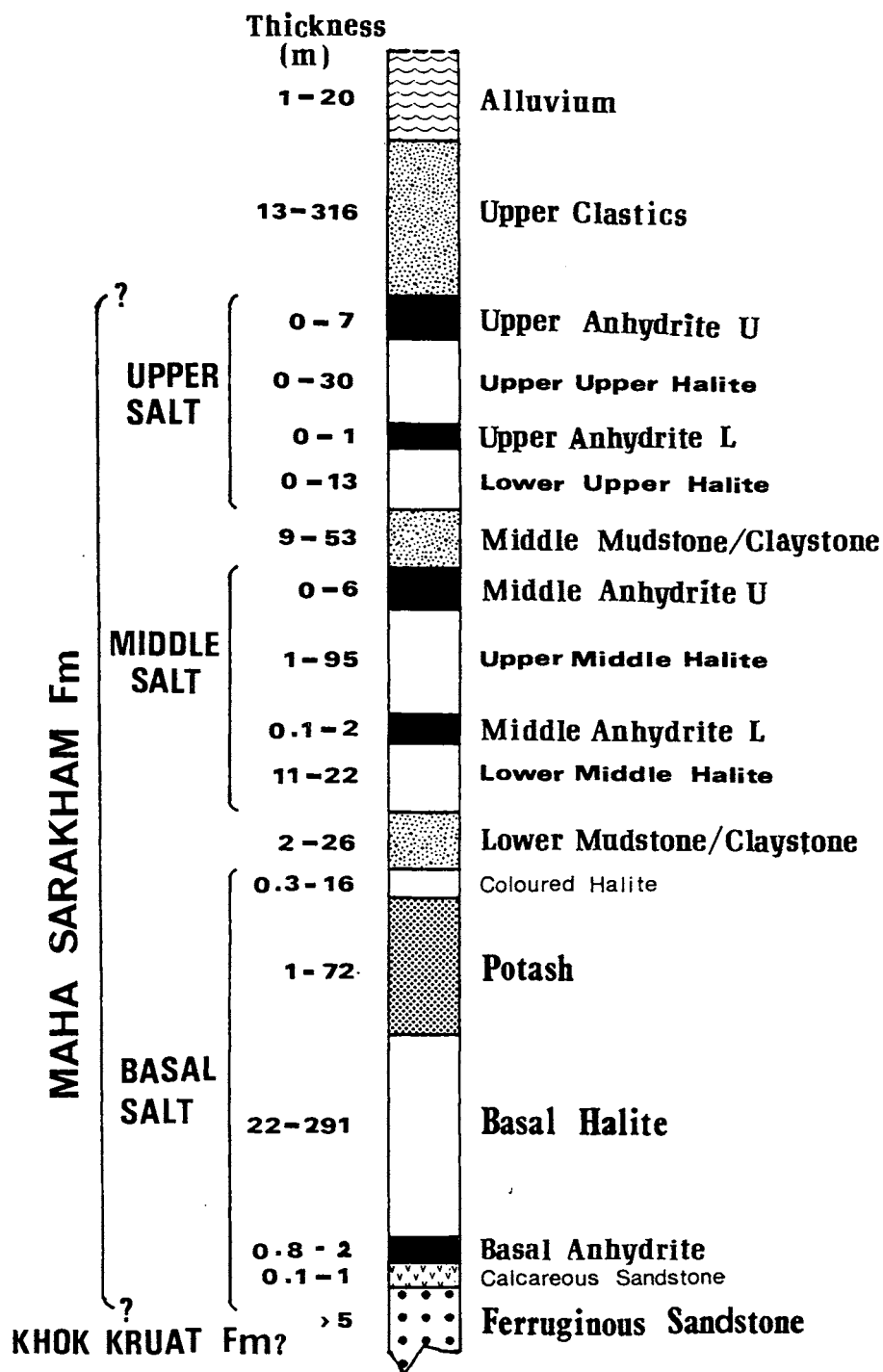


Fig. 2. Type I sedimentary sequence of the Maha Sarakham evaporite deposits in Bamnet Narong area.

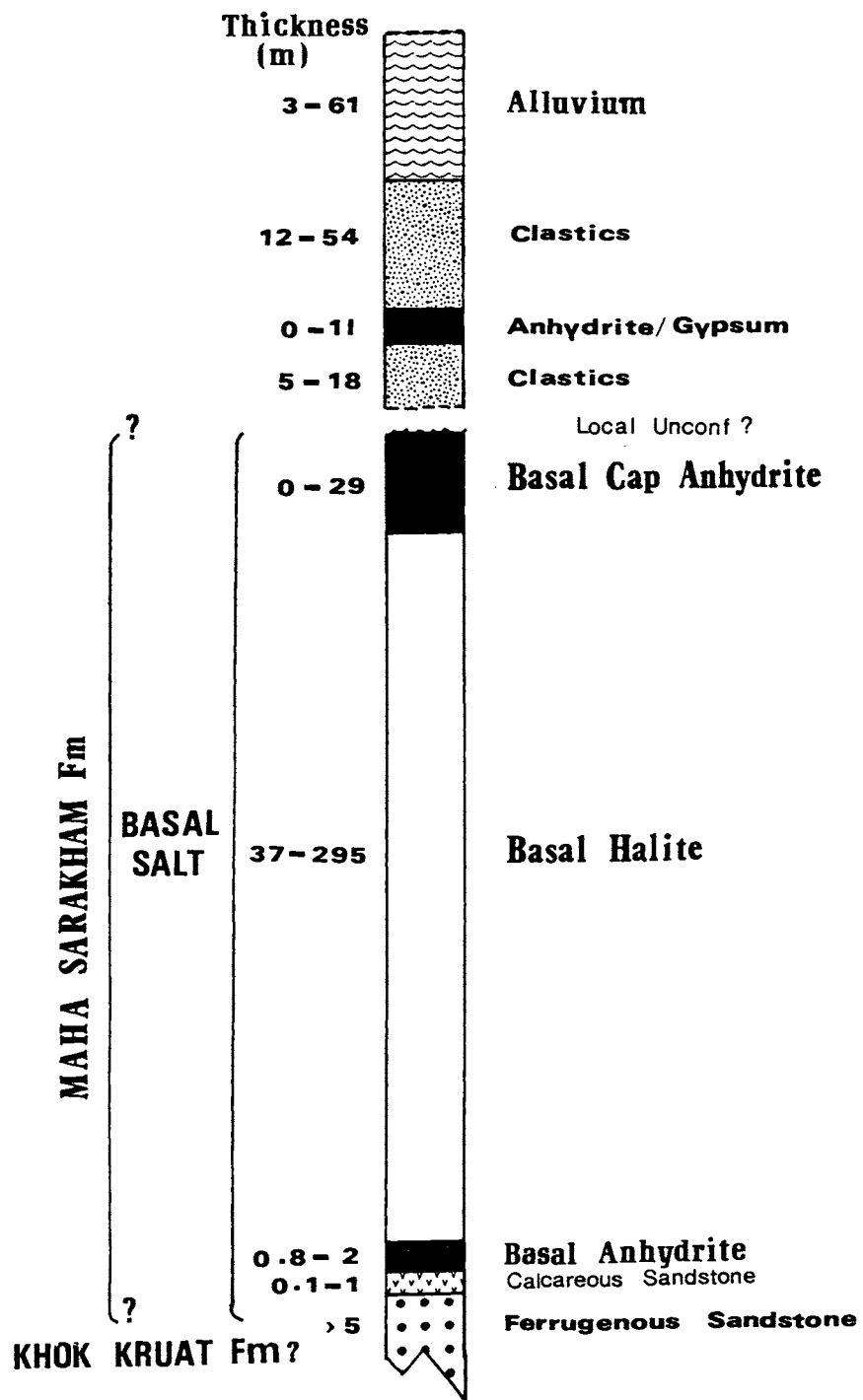


Fig. 3. Type II sedimentary sequence of the Maha Sarakham evaporite deposits in Bamnet Narong area.

SAMPLING ANALYTICAL PROCEDURE

A total of 10 anhydrite samples were analyzed for the $\delta^{18}O$ and $\delta^{34}S$ values in this study. From each anhydrite bed, except the Upper Anhydrite L, two representative samples, one from the lower part and the other from the upper part, were taken for the analysis. The Upper Anhydrite L bed was not analysed because of the scarcity of the sample recovery. All of these samples were obtained from four selected drill cores in the Bamnent Narong area with the maximum distance between the drill holes being approximately 6 km apart. These anhydrite samples were also analyzed for some trace elements (i.e., Mn, Fe, Sr and Ba; Yumuang 1983).

The sulphur and oxygen isotopic analyses were performed at the Institute for Thermal Spring Research of Okayama University in Japan. The anhydrite samples were dissolved, purified and subsequently converted to $BaSO_4$ according to the procedure by Kusakabe and Chiba (1983). The graphite reduction method (Mizutani, 1971; Sakai and Krouse, 1971) was used to obtain CO_2 gas from the $BaSO_4$ for oxygen isotopic analysis. The oxygen isotopic analysis was performed on the VG Microsmass 903 equipped with double inlet system and triple collectors. The SO_2 gas was obtained by the thermal decomposition of $BaSO_4$ (Yanagisawa and Sakai, 1983) and was used for sulphur isotopic analysis. The sulphur isotopic analysis was performed on the Hitachi RMU-6RS equipped with double inlet system and double collectors.

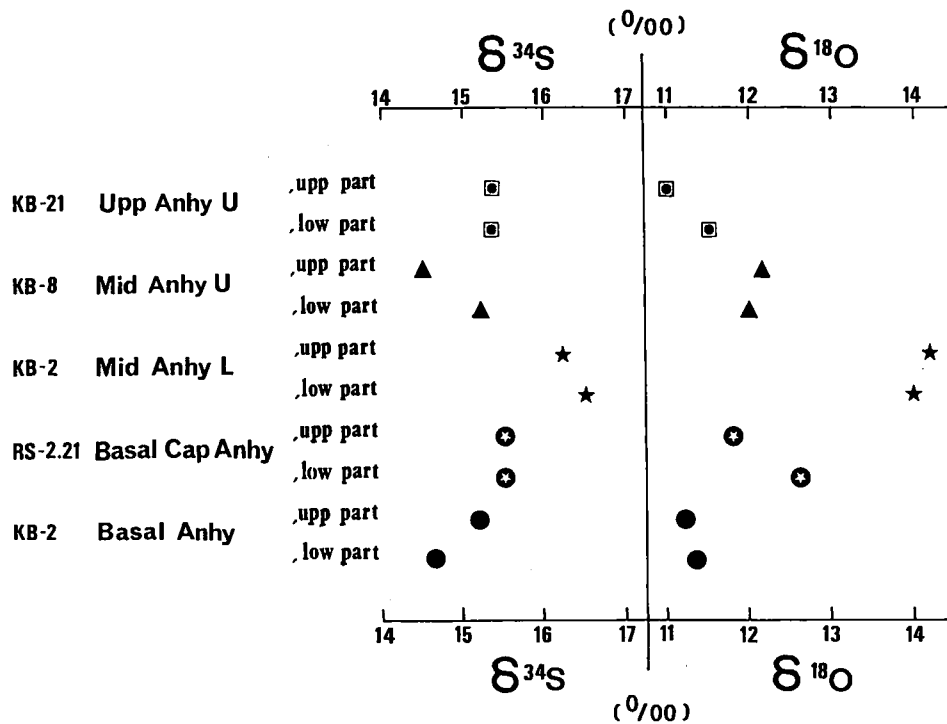


Fig. 4. Comparison of sulphur and oxygen isotopic values of the Maha Sarakham anhydrite from stratigraphically lower to upper part of the sedimentary sequence.

The $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values of anhydrite are reported relative to CDT and SMOW standards, respectively. The analytical uncertainties are $\pm 0.2\%$ for sulphur isotope and $\pm 0.3\%$ for oxygen isotope. The $\alpha_{\text{CO}_2-\text{H}_2\text{O}}$ value of 1.0412 at 25°C (O'Neil *et al.*, 1975) was used in the calculation of the $\delta^{18}\text{O}$ values.

RESULTS

The results of sulphur and oxygen isotopic analysis of the Maha Sarakham anhydrite from the Bamnet Narong area is tabulated in Table 1 and graphically displayed in Fig. 4 from the stratigraphically lower to upper parts of the sedimentary sequences. The $\delta^{34}\text{S}$ values of anhydrite vary within a narrow range from 14.5 to 16.5‰ with an average of 15.4‰ whereas the $\delta^{18}\text{O}$ values are relatively more spreading from 11.0 to 14.2‰ with an average of 12.2‰. The $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values of anhydrite within a single bed show very small variations. There seems to be no systematic variation in both $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values with respect to their stratigraphic position. There was also no distinct difference in both $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values between the Basal Cap Anhydrite and those of the Normal anhydrite. The results were also plotted in a $\delta^{18}\text{O}$ versus $\delta^{34}\text{S}$ diagram (Fig. 5). Again no obvious trend can be observed except for those of the Middle Anhydrite L bed show a slightly heavier $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values relative to the remaining values (Fig. 5).

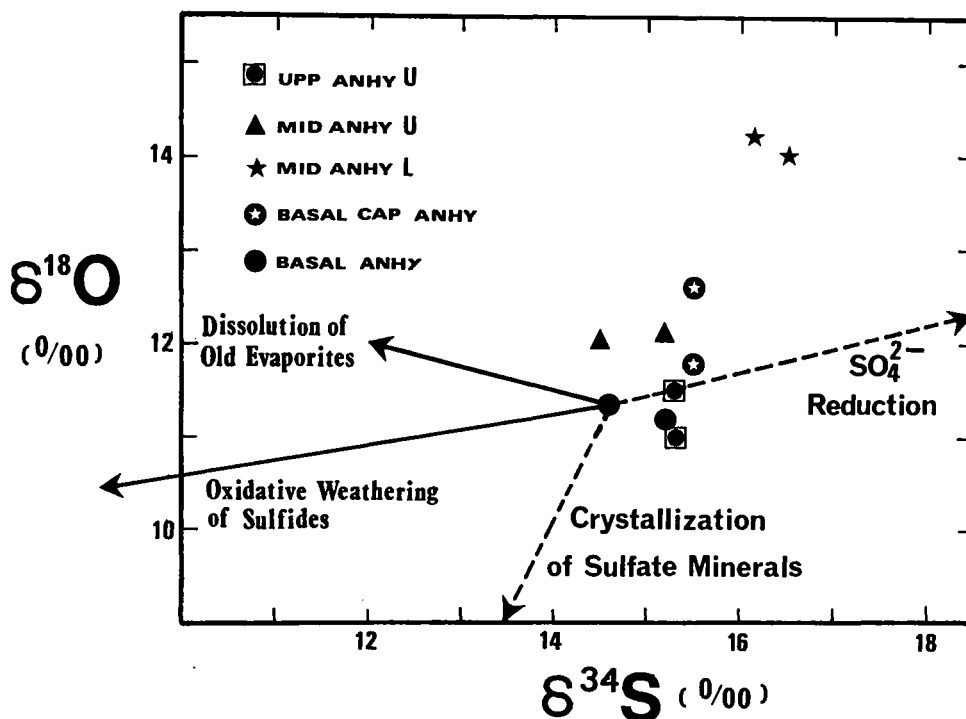


Fig. 5. Comparison of sulphur versus oxygen isotopic values of the Maha Sarakham anhydrite. See text for details of the four vectors.

TABLE I
SULPHUR AND OXYGEN ISOTOPES OF MAHA SARAKHAM ANHYDRITE

No.	Depth (m)	Sample	$\delta^{34}\text{S}$ (‰)	$\delta^{18}\text{O}$ (‰)
KB-2	354.27–364.31	Basal Anhydrite (lower part)	11.3	14.6
KB-2	353.98–354.02	Basal Anhydrite (upper part)	11.2	15.2
RS-2.21	66.97–67.07	Basal Cap Anhydrite (lower part)	12.6	15.5
RS-2.21	66.00–66.13	Basal Cap Anhydrite (upper part)	11.8	15.5
KB-2	100.93–100.98	Middle Anhydrite L (lower part)	14.0	16.5
KB-2	100.67–100.70	Middle Anhydrite L (upper part)	14.2	16.2
KB-8	57.15–57.27	Middle Anhydrite U (lower part)	12.1	15.2
KB-8	55.14–55.29	Middle Anhydrite U (upper part)	12.0	14.5
KB-21	203.35–203.51	Upper Anhydrite U (lower part)	11.5	15.3
KB-21	170.35–170.50	Upper Anhydrite U (upper part)	11.0	15.3

DISCUSSION

Post-Depositional Isotopic Modification

The Maha Sarakham Formation was deposited in a rather continuous fashion on the relatively stable Khorat platform. The overlying semi-consolidated and loose sediments are also rather thin (approximately 300 meter thick in the Bamnet Narong area; Yumuang, 1983). The evaporite beds should not have therefore undergone any significant degree of metamorphism. The processes that may have taken place during and after the deposition of the evaporitic sequences are recrystallization of some minerals, dehydration, flowage of salt and expulsion of interstitial water. The above processes should not have any effect on the sulfur isotopic composition of the original sulphate minerals. Therefore, the $\delta^{34}\text{S}$ values of those anhydrite should reflect their original isotopic compositions.

In terms of oxygen isotope, however, the dehydration of sediments or hydrous minerals may cause the isotopic re-equilibration between the $\delta^{18}\text{O}$ of pore water or dehydrated water and those of the anhydrite during the rise in temperature of burial metamorphism. If we assume a normal geothermal gradient of $25^\circ\text{C}/\text{km}$ in this area, the temperature at 300 meter depth of sediments would rise by only 8°C . This small increase in temperature should have negligible effect on the original $\delta^{18}\text{O}$ values of sulfate minerals. Besides, according to experimental work by Chiba *et al.* (1981), the oxygen isotopic exchange rate between water and sulfate is extremely slow at temperature below 200°C .

Hite (1982) proposed one important post-depositional change, that of extensive dissolution of the evaporite beds particularly in the area of salt anticline. He suggested that the salt anticline had a genetic relation to the incision of old stream channels during the Pleistocene age (?). These old stream channels would supply large amounts of water to dissolve away all the soluble salts (i.e., halite, carnallite, sylvite, tachyhydrite) and leave relatively insoluble anhydrite as a residue cap on top of the salt anticline (Hite, 1982; Yumuang, 1983).

Even though a large amount of groundwater may possibly be in contact with those anhydrites especially the Basal Cap anhydrite, there should be no significant degree of oxygen isotopic exchange between the groundwater and these anhydrites. This is because the rate of oxygen isotopic exchange between water and sulfates at normal groundwater temperature is extremely slow (Chiba *et al.*, 1981). Therefore, the $\delta^{18}O$ values of all those anhydrite should be their original isotopic compositions.

The Isotopic Composition of the Khorat Seawater Sulfate

The $\delta^{34}S$ and $\delta^{18}O$ values of the anhydrite are likely to represent their original isotopic composition of sulfate minerals that precipitated in equilibrium with seawater sulphate from the restricted Khorat sea. We still do not know whether the Khorat seawater sulphate has the same isotopic value as the world ocean sulphate during that geologic time or whether the isotopic composition of Khorat seawater sulphate was modified by local input and output of sulphur and oxygen through the sedimentary sulphur cycle. It is also uncertain whether these values are distinctly different from those of the world ocean sulphate.

The isotope geochemistry of sulphur and oxygen in sedimentary sulphur cycle has been described repeatedly and well summarized by Claypool *et al.* (1980). The possible and well-accepted geological factors that may control the geochemical input and output of sulphur and oxygen into the Khorat sea are 1) dissolution of pre-existing evaporites and their addition to the Khorat sea, 2) oxidative weathering of old sulphides (mainly as pyrite from shale) and feeding back to the sea as sulphate, 3) crystallization of sulphate minerals as gypsum or anhydrite in the evaporite sequences, 4) reduction of seawater sulphate by bacteria and transforming sulphate oxygen into CO_2 gas and sulphate sulphur to sulphides and later fixed as pyrite in muds.

The factor of dissolution of pre-existing evaporites is less certain. This is because there is no report of large occurrences of anhydrite or gypsum deposits in the pre-Maha Sarakham Formation of the Khorat Group. However, large gypsum and anhydrite deposits were reported in the Pichit-Nakorn Sawan area (approximately 1.1 million ton measured reserve) on the west of the Khorat Plateau and in the Loei area (approximately 0.5 million ton inferred reserve) on the northwest of the Khorat Plateau (Yeamniyom, 1977). According to Gardner (1967), the Pichit-Nakorn Sawan anhydrite-gypsum deposits rested unconformably over the Permian rocks and is believed to be of the same age as the Maha Sarakham Formation. The Loei anhydrite-gypsum deposits, however, were found to interbedded with the Upper Paleozoic rocks of Kanchanaburi Group (Yeamniyom, 1977).

If we assume that the Loei anhydrite-gypsum was the older, then sulphate

minerals could be dissolved into the Khorat sea. Using the sulphur and oxygen isotope age curves of the world evaporites summarized by Claypool *et al.* (1980), the Loei anhydrite-gypsum can be estimated to have values around 12‰ for $\delta^{34}\text{S}$ and 12‰ for the $\delta^{18}\text{O}$. By progressive dissolution of old sulphate minerals into the Khorat sea, the value of the Khorat seawater sulphate should continuously change toward the values of those older sulphates.

The pre-Maha Sarakham Formation in northeastern Thailand are predominantly continental red-bed facies of the Khorat Mesozoic Group. The contribution of sulphate from the weathering of old sulphides in those rocks is therefore rather limited. However, there may be some contribution of sulphate from the oxidation of sulphides in the marine facies from the western part of the Khorat Plateau.

The $\delta^{34}\text{S}$ values of sedimentary sulphides are rather wide in range (–70 to +70‰, Ohmoto and Rye, 1979) but are generally enriched in the light isotope. If we accept the mean values estimated by Claypool *et al.* (1980) of –17‰, the oxidation of these sulphides would produce sulphates of approximately the same sulphur isotopic values as those of sulphides (Ohmoto and Rye, 1979). In cases of the oxygen isotope of the sulphate produced from the oxidation of sulphides, however, the $\delta^{18}\text{O}$ values of sulphate product should be the mixture of fractionated air oxygen and meteoric water oxygen and could be estimated to be around +2‰ (Claypool *et al.*, 1980). Hence, by progressive addition of the light sulphates from the weathering of old sulphide into the Khorat sea, the Khorat seawater sulphate would be depleted isotopically toward the sulphide values.

In the process of crystallization of gypsum or anhydrite in the evaporite sequence, the fractionation factors between sulphate minerals and seawater sulphate were approximated to be 1.65‰ for $\delta^{34}\text{S}$ and 3.5‰ for $\delta^{18}\text{O}$ (Thode and Monster, 1965; Claypool *et al.*, 1980). This process would therefore produce sulphate minerals that became progressively enriched in the heavy isotope within the restricted Khorat basin.

The last factor, the bacterial reduction of seawater sulphate, is poorly known. However, according to Claypool *et al.* (1980), the best estimated fractionation factor between the produced sulphide and the remaining sulphate is –4‰ for $\delta^{34}\text{S}$ and between CO_2 and the sulphate is +10‰ for $\delta^{18}\text{O}$. In other words, the seawater sulphate will be progressively heavier in both $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values as bacteria continued to reduce more seawater sulphate to sulphide and CO_2 gas in the Khorat sea.

By assuming that the Basal Anhydrite from the lower part was the first fraction of sulphate mineral precipitated from the Khorat seawater, it is possible to draw vectors of all four factors from this point (Fig. 5) using appropriate fractionation factors and isotopic values mentioned earlier to test if any of these factors were dominant in the Khorat sea.

As shown in Fig. 5, the isotopic values of those anhydrite are rather scattered and do not follow any of those vectors. The lack of significant trend in this plot suggests that those four known factors were not operating in the Khorat sea. Furthermore, the constancy of $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values within a single anhydrite bed suggests that these known factors surely were not significant in each evaporitic cycle. The intrabasin

dispersion of the $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values of those sulphate minerals may be caused by other unknown factors. However, because of the fact that the variation of those values is rather small, it is reasonable to conclude that the Khorat seawater sulphate was not modified isotopically to any significant degree from those of the world ocean sulphate by any other factors during that geologic time. In other words, the $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values of Maha Sarakham anhydrite should represent those of sulphate minerals precipitated in equilibrium with the world ocean sulphate during that date.

Age of the Maha Sarakham Evaporite

The age of the Maha Sarakham Formation is highly controversial. Its age was mostly inferred from the stratigraphic succession (e.g. Sattayarak, 1983); with a few evidences obtained from the studies of palaeomagnetism (Maranate, 1982) and of pollens and spores (quoted in Sundharovat, 1978). Their ages were considered to vary from as old as Lower Cretaceous (Gardner *et al.*, 1967) to as young as Pleistocene (Sundharovat, 1976). Hence, its definitive age is still uncertain.

Sulphur isotopes of marine evaporites are known to vary with age and are well summarized by Claypool *et al.* (1980). Theoretically, by knowing the isotopic data of a marine a evaporite alone, it is not possible to use the sulphur isotopic age curve to tell the approximate age of that anhydrite because of the non-lineament of the curve with respect to time. Several portions of the curve also have very few data base. Fortunately,

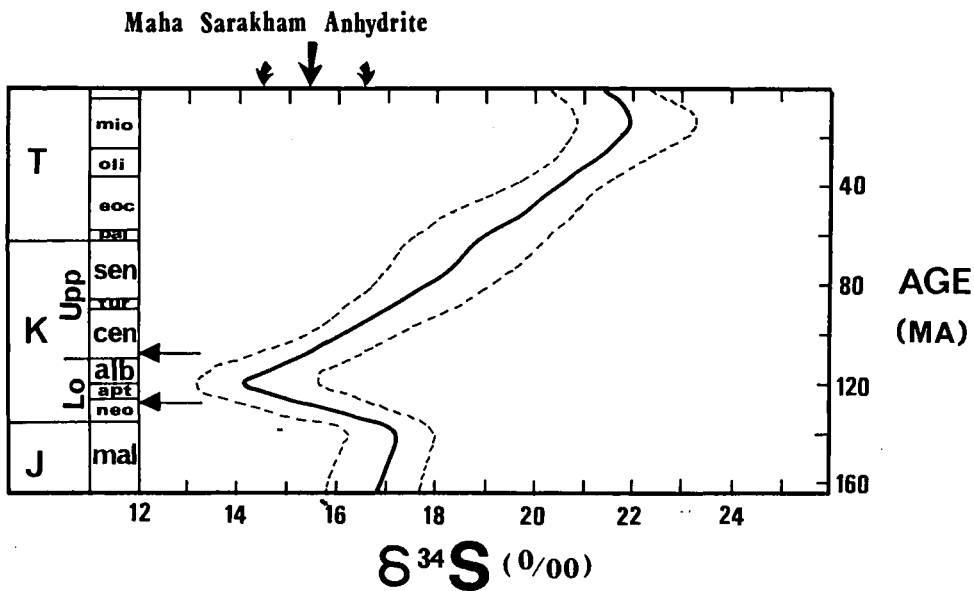


Fig. 6. Estimation of the age of Maha Sarakham evaporite from Bamnet Narong area. The sulphur isotope age curve of marine evaporites is taken from the upper portion of the sulphur curve revised by Claypool *et al.* (1980). The heavy and dash lines are the best estimate and uncertainties of the curve respectively (Claypool *et al.*, 1980). The large arrow above the diagram is the average $\delta^{34}\text{S}$ value of the Maha Sarakham anhydrite and the small arrows nearby are the spread of the values. Two horizontal arrows in the diagram are the corresponding age of the average $\delta^{34}\text{S}$ value. See text for further discussion.

because the underlying Khok Kruat Formation is Early Cretaceous or older (Buffetaut and Ingavat, 1985; Marante, 1982), the age of Maha Sarakham evaporite should be somewhere in Cretaceous or younger. Hence, we are dealing with the upper portion of the sulphur age curve which seems to be somewhat linearly shifted with respect to time (Fig. 6). The data base used to construct this portion of the curve is fairly well established. Therefore using the sulphur isotopic data of the Maha Sarakham anhydrite and the upper portion of the sulphur age curve of the world marine evaporites, it is possible, as an independent means, to put some limits on the age of Maha Sarakham evaporite in the Bamnet Narong area.

Using the average $\delta^{34}\text{S}$ value of the Maha Sarakham anhydrite of 15.4‰, the corresponding ages are unfortunately located at either Cenomanian or Neocomian because the curve turns at around middle Cretaceous (Fig. 6). If we have to choose between these two ages, the Cenomanian is our best choice because the underlying Khok Kruat Formation was reported to have an age from Jurassic to middle Cretaceous by palaeomagnetic study (Marante, 1982) or at Abtian, Albian (upper part of Early Cretaceous) by using fossil vertebrates (Buffetaut and Ingavat, 1986).

By taking the maximum uncertainties of the isotopic values and of the age curve, the lower limit of the Maha Sarakham evaporite could be as old as Jurassic and the upper limit, Turonian (Fig. 6). Hence, the age of the Maha Sarakham evaporite at

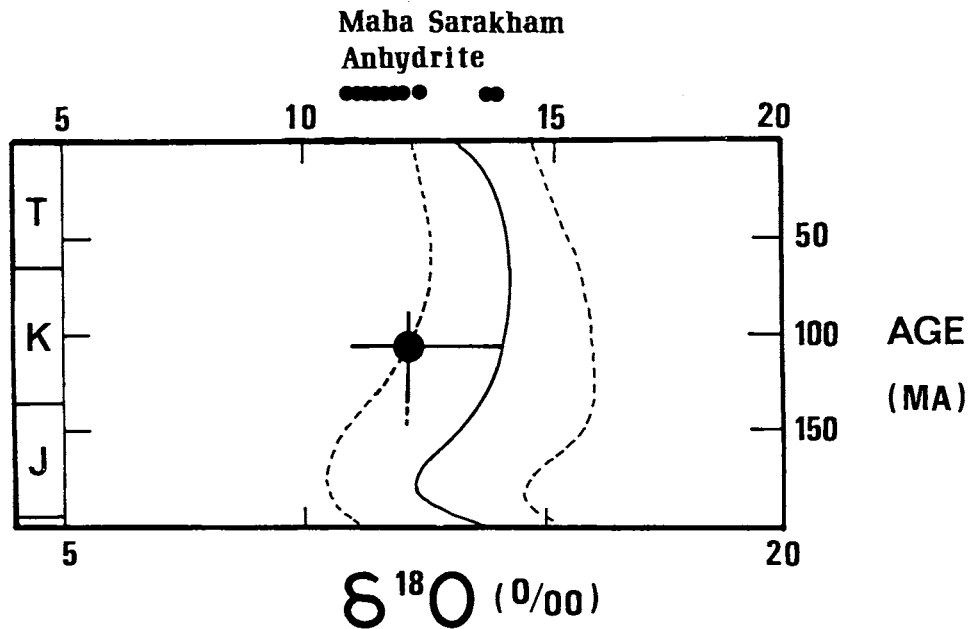


Fig. 7. Comparison of the $\delta^{18}\text{O}$ values of anhydrite and that of the best estimate by Claypool *et al.* (1980). The oxygen isotope age curve of the marine evaporite is taken from the upper portion of the oxygen curve revised by Claypool *et al.* (1980). The heavy and dashed lines are the best estimate and uncertainties respectively (Claypool *et al.*, 1980). The solid circle is the average $\delta^{18}\text{O}$ values and the best estimated age from Figure 6. The horizontal and vertical lines represent uncertainties. See text for further discussion.

Bamnet Narong area should not be younger than Turonian (Middle of Upper Cretaceous).

In fact, there was a report on the study of spores pollens collected from a single drill core which penetrated through the Maha Sakham Formation by Exxon Co. (quoted in Sundharovat, 1978). The age of the Formation varies from Jurassic to Miocene and the best estimate was at Albiam/Cenomanian (middle Cretaceous). This is essentially similar to the isotope ages.

The upper portion of the oxygen isotope age curve of the world marine evaporite summarized by Claypool *et al.* (1980) is reproduced as Fig. 7. As can be seen, the curve shows a rather small variation and large uncertainties in the Cretaceous and Tertiary. If we plot the average $\delta^{18}O$ value and uncertainties of the Maha Sakham evaporite and their ages, estimated from the sulphur isotope data on the oxygen age curve, it is rather obvious that the data is slightly off from the best estimated curve (Fig. 7). Because the data base to construct the portion of this curve especially in Cretaceous is scarce (Claypool *et al.*, 1980), we believe that our data should be added to the list for future revision of the oxygen age curve.

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