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Measurement of crystallinity and relative amount of clay minerals in the Kathmandu Basin sediments by decomposition of XRD patterns (profile fitting)

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ABSTRACT

The estimation of crystallinity and relative amount of clay minerals in the Kathmandu Basin sediments were made by using decomposition procedure of X-ray diffraction (XRD) patterns. Based on this study an attempt has been made to reconstruct the palaeoclimatic variation recorded in the clay minerals. The variations of the two illite crystallinity indices, Lanson index (LI) and modified Lanson index (MLI) of the drill-core sediments between 5 m and 49 m depth are linked to those of the relative amount of smectite to illitic minerals or to chlorite. The relative amount of kaolinite to illitic minerals or to chlorite roughly showed mirror image of the relative amount of smectite to illitic minerals or chlorite, respectively.

The variation of the hydrolysis condition inferred from these indices were congruous with the variation of $\delta^{18}O$ from planktonic foraminifers of deep sea sediments in the Arabian Sea, confirming that the climatic variations during the last ~ 450 kyr in the Kathmandu Basin were closely related to global climate, without apparent local climatic changes. In addition, they depicted that it was dry during the cold climate and wet during the warm climate in the Kathmandu Basin.

INTRODUCTION

Presently, recent climatic history of the Earth is being intensively investigated in the world. Especially, the origin and change of Asian monsoon are extremely important while considering the climatic variations of the Earth in the past and future. The Asian monsoon system and its evolution are known to be closely linked to the Himalayan-Tibetan orogen (*e.g.* Ruddiman 1997). Intermontane basins and peripheral basins in and around the Himalayan Range and Tibetan Plateau must have preserved a good record of past climate variations and tectonic events in the orogen. The Kathmandu Basin is the best target for clarifying the variability of the Asian monsoon climate and its linkage to the uplifting of the Himalayan-Tibetan orogen, since it is located on the southern slopes of the central Himalayas and filled with thick sediments from late Pliocene to Quaternary (Sakai et al. 2000, 2001; Fujii and Sakai 2001). Therefore, our group initiated an academic drilling project "Paleo-Kathmandu Lake (PKL) Project" in the Kathmandu Basin (Sakai et al. 2000) with H. Sakai as the leader, and has alrready carried out drillings at five sites in the basin.

Prior to the PKL project, our group had carried out the preliminary investigation on fossil pollens in the Kathmandu Basin sediments and the characteristics of the sediments using a series of core samples (JW-3), which were previously drilled for the ground water production (Sakai et al. 2000, 2001; Fujii and Sakai 2001). We also investigated the clay minerals in the same samples to obtain the palaeoclimatic information recorded in the Kathmandu Basin sediments. Clay minerals are one of useful indicators of palaeoclimate, since they are formed through weathering or hydrolysis processes in geological history and basically express the intensity of weathering or hydrolysis in the land masses adjacent to sedimentary basin (Chamley 1989). The information provided from such clay minerals fundamentally integrates the combined effects of temperature and precipitation.

Practically, the climatic interpretation of clay mineral study is based on the variations of the illite crystallinity index and/or the relative amount of alteration minerals to primary minerals (*e.g.* illite and smectite). Particularly, the Kübler Index (KI) (Kübler 1964), which is a conventional illite crystallinity index defined by the full width at half maximum intensity (FWHM) of 10 Å illite X-ray diffraction (XRD) peak, has extensively been used to reconstruct the palaeoclimate (*e.g.* Chamley 1989; Fukuzawa et al. 1997; Lamy et al. 2000). However, it has been reported that KI is significantly larger for finer fractions (Srodon 1979; Srodon and Eberl 1984) and that Lanson illite crystallinity index (LI) (Lanson 1997), which accounts for the relative proportion of illite crystallites with low coherent scattering domain size (CSDS), is more available for the estimation of illite crystallinity than KI (Kuwahara et al. 2001). LI can be determined by use of the characteristics of three elementary peaks obtained by decomposing the complex XRD bands near 10 Å using the three elementary peaks corresponding to three mineral phases with different illite content and mean CSDS thickness (Lanson 1997).

We have accurately measured the illite crystallinity and relative amount of alteration minerals to primary minerals by the decomposition procedure of XRD patterns of clay minerals. In this paper, we introduce the details of our decomposition procedure and report the availability of that for the reconstruction of palaeoclimatic variation.

ILLITE CRYSTALLINITY OR ILLITE CRYSTALLITE THICKNESS BY XRD

It is known that the crystallinity of illite can be used to characterize the palaeoclimates, since it represents quantitative indicators of hydrolyses processes on land, which closely relate to precipitation and temperature (Chamely 1989). In particular, KI has extensively been used to reconstruct the palaeoclimatic variations, as described above. KI is defined by FWHM of illite 001 (for 1*M*) XRD peak (of course, 002 for 2M_1) (Fig. 1). It was believed that the FWHM of illite 001 for 1*M* and/or 002 for 2M_1 XRD peak near 10 Å is inversely proportional to the crystallinity and structure order of illite particles (Srodon and Eberl 1984; Chamely 1989). However, Srodon (1979) and Srodon and Eberl (1984) suggested that FWHM of the 10 Å illite XRD peak is mostly a function of the amount and composition of the illite-smectite mixed layer minerals (I-S) of the sample. Thus, KI is significantly larger for finer particle fraction, because the I-S has a finer particle size than illite (Srodon and Eberl 1984).

There are two equally valid ways to describe illitic crystals composed of illite and smectite (Eberl and Blum 1993). Fig. 2 shows I-S represented as a MacEwan crystallite and as an aggregate of fundamental particles. From the MacEwan crystallite viewpoint, such mixed layers are composed of interstratified illite and smectite layers (Eberl and Blum 1993). The percentage of each can be determined from XRD peak positions for glycol-solvated samples (Srodon and Eberl 1984; Moore and Reynolds 1989). On the other hand, according to the theory of interparticle diffraction (Nadeau et al. 1984), I-S is composed of discrete fundamental illite particles that absorb water on their basal *X-Y* surfaces. Smectite layers are water-rich interfaces that diffract coherently along *Z* with adjacent illite particles. Therefore, the formation of such smectite layers by mechanical mixing should largely occur in very thin illitic particles. This may closely relate to that KI is significantly larger for finer particle fraction, as mentioned above. Very recently, Kasama et al. (2001) reported that the mixtures of smectite and rectorite (ordered (R1) I-S containing 50% illite) of the fractions under 0.1 μm show interparticle diffraction, whereas the mixtures of them of 0.5 – 2.0 μm fractions do not show interparticle diffraction.

Lanson and co-workers demonstrated that the complex XRD bands near 10 Å show not only a difference in FWHM (that is, KI) but also a shift in peak position, associated with mean values of illite content and/or CSDS for a given illitic particle population (Lanson and Besson 1992; Lanson and Velde 1992; Lanson 1997) (Fig. 3). CSDS is equivalent to crystallite size but may or may not be equal to fundamental particle thickness, because each fundamental particle may or may not diffract coherently (Eberl and Blum 1994). Lanson (1997) also showed that the complex XRD peak near 10 Å of

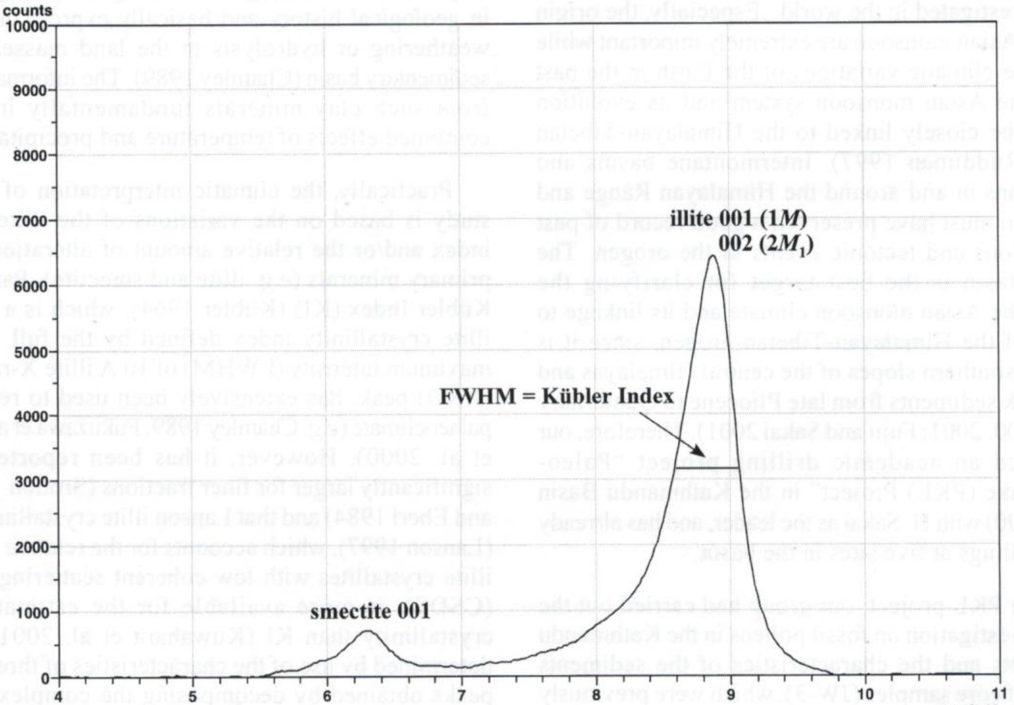

Fig. 1: XRD pattern of mixture of illite and smectite (oriented sample). The full width at half maximum intensity (FWHM) of illite 001 peak (for 1*M*) (and/or 002 for 2M_1) (that is, Kübler Index) is also shown.

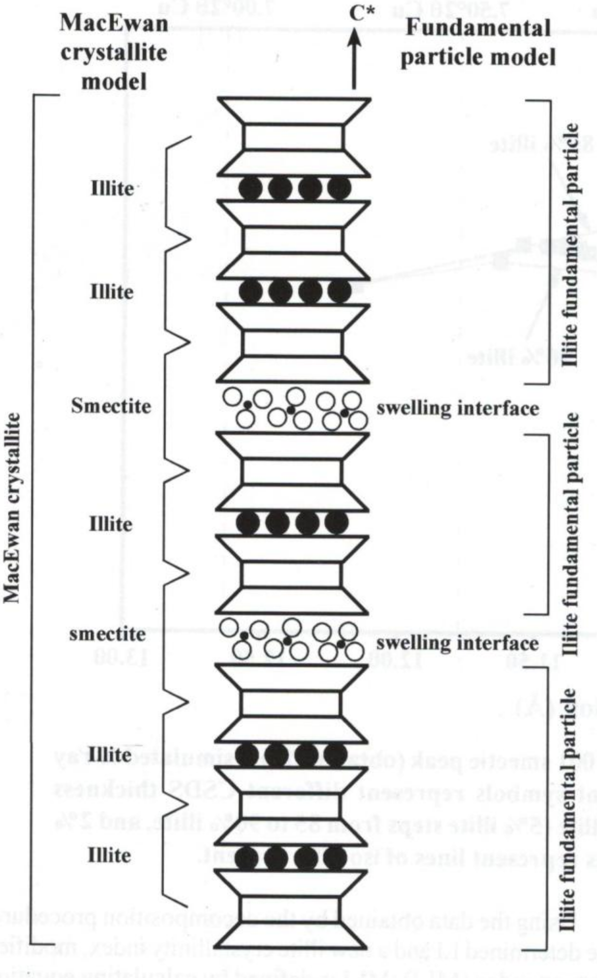

Fig. 2: Illite-smectite mixed layer minerals (I-S) represented as a MacEwan crystallite and as an aggregate of fundamental particles. Anvils represent 2:1 phyllosilicate layers. Large dark circles are fixed K, open circles are water molecules, and dark circles are exchangeable cations. MacEwan crystallitie thickness is equivalent to coherent scattering domain size (CSDS) thickness.

natural illitic minerals are generally observed to be asymmetrical (Fig. 1) and that the asymmetry of this peak is in fact caused by the CSDS distribution rather than an artifact of the experimental equipments. He, therefore, decomposed the complex XRD bands near 10 Å using three elementary peaks corresponding to three different phases with different illite content and different mean CSDS thickness, that is, I-S, poorly crystallized illite (PCI), and well-crystallized illite (WCI) (Fig. 4). Once the decomposition procedure is done for an XRD pattern, the characteristics (*e.g.* peak position, FWHM, and intensity (peak area)) of each elementary peak are accurately determined. From the characteristics of the elementary peaks representing a phase, the mean values of some physico-chemical characteristics such as composition and CSDS are derived (Lanson 1997).

Then, Lanson (1997) proposed a new illite crystallinity index, LI, which is expressed as:

$$LI = 0.1/[\text{PCI peak relative intensity} \times \text{PCI peak FWHM} \times (\text{PCI peak position} - \text{WCI peak position})] \quad (1)$$

where:

$$\text{PCI peak relative intensity} = \text{PCI intensity} / (\text{PCI intensity} + \text{WCI intensity} + \text{I-S intensity}). \quad (2)$$

Here, peak FWHM is expressed in °2θ CuKα and peak position is expressed in Å. Higher the LI, higher the illite crystallinity, or smaller KI. LI accounts for the relative population of illite crystallites with low CSDS, that is, PCI, which influences on both peak position and FWHM. LI has been applied to the evolution of illite crystallinity during the diagenetic processes (Lanson 1997) and has been reported to be more available for the estimation of illite crystallinity than KI (Kuwahara et al. 2001).

EXPERIMENTAL DETAILS

The samples used are the drill-core JW-3 which is located on the bank of the Bagmati River at Sundarighat in the west central part of the Kathmandu Basin. The core samples consist of 189 pieces of slime collected at one metre intervals from the 284.3 m deep drill-well. The samples from JW-3 are the identical ones as studied by Fujii and Sakai (2001), and the details of the drilled sediments have been described by Fujii and Sakai (2001). In this study, 43 samples of the upper part of the drill-well from 5 m to 49 m in depth, which are organic black or dark gray mud called "Kalimati Clay", have been examined by XRD and the decomposition procedure. According to Fujii and Sakai (2001), the mean sedimentation rate of the upper 115 m of JW-3 is about 104 mm/kyr and that the base of the drill-well is probably older than 2.6 Ma.

Sample preparation was performed according to Kuwahara et al. (2001). Firstly, we separated the clay fraction under 2μm from each slime sample by gravity sedimentation. Then, about 200 mg of this fraction was collected by the Millipore® filter transfer method to provide an optimal orientation (Moore and Reynolds 1989). The filter used is the Gelman® GA-9, 0.45 μm pore, 47 mm diameter Metricel© filter. The thickness of a clay cake formed on the filter is over 15 mg/cm², which is adequate for XRD quantitative analysis (Moore and Reynolds 1989). The clay cake was then transferred onto a glass slide. Both air-dried (AD) and ethylene glycol solvated (EG) preparations were done for each sample. The EG preparation was carried out to expose the sample to the vapor of the reagent in desiccator over 8 hr at 60 °C.

All XRD measurements were performed on a Rigaku X-ray Diffractmeter RINT 2100V, using CuKα radiation monochromatized by a curved graphite crystal in a step of 0.02° with a step-counting time of 4 seconds.

The decomposition of the obtained XRD patterns was performed with an Apple Power Macintosh computer and a scientific graphical analysis program XRD MacDiff

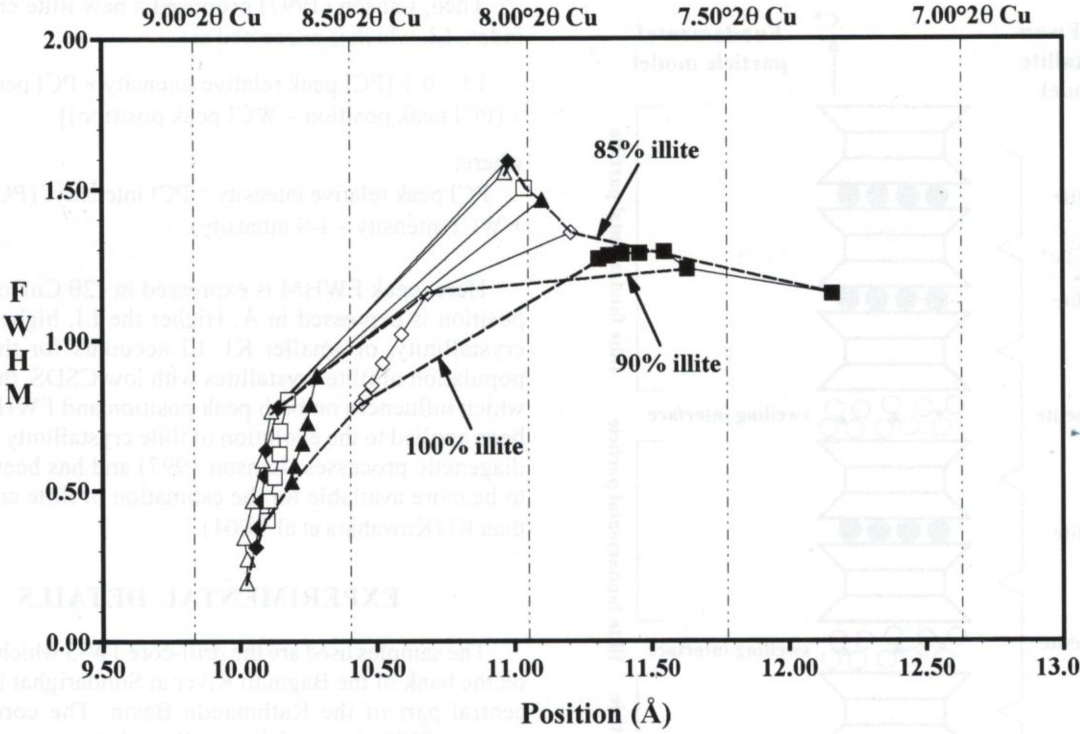

Fig. 3: Identification diagram for ordered I-S, 001 illite / 001 smectie peak (obtained from simulated X-ray patterns) (modified from Lanson 1997). The different symbols represent different CSDS thickness distributions. I-S composition varies from 85 to 100% illite (5% illite steps from 85 to 90% illite, and 2% illite increments from 90% to 100% illite). Dashed lines represent lines of iso illite content.

(Petschick 2000), according to Lanson (1997). First, a raw file was treated with a preliminary smoothing to decrease the effect of statistical counting errors, and then a background was subtracted to eliminate most of its contribution to the peaks (Fig. 5a). Finally, the elementary peak fitting was done (Fig. 5b). All decompositions were done with symmetrical elementary peaks with Gaussian shape.

The representative XRD patterns of AD and EG samples in the drilled sediments are shown in Fig. 6. Clay mineral assemblage of these sediments consists of illitic minerals ($2M_1$) (PCI and WCI), I-S, smectite, chlorite, and kaolinite. Therefore, we decomposed the 3°-11° 2θ CuKα region of both AD and EG samples using five elementary peaks which correspond to smectite, chlorite, I-S, PCI and WCI, to determine the illite crystallinity and the relative amount of smectite to illitic minerals or to chlorite. The 23°–27° 2θ CuKα region of AD samples was also decomposed to separate kaolinite and chlorite using two elementary peaks corrensponding to them. The decomposition procedure can accurately determine the characteristics (*e.g.* peak position, FWHM, and intensity (peak area)) of each elementary peak (Fig. 5b). Kuwahara et al. (2001) have shown that the error in our method is very small, which are sufficient for the reproducibility of results, by both repeating four times the decomposition for an XRD pattern of each sample and performing the decomposition for four XRD patterns collected for a given sample.

Using the data obtained by the decomposition procedure, we determined LI and a new illite crystallinity index, modified Lanson index (MLI). MLI is defined by calculating equation (1) with equation (3) instead of equation (2),

PCI peak relative intensity = PCI intensity / (PCI intensity + WCI intensity) (3),

and estimates the illite crystallinity only from the difference between PCI and WCI. MLI is likely more reliable for these samples, since the standard deviations on peak position and FWHM of an I-S peak tend to be larger than those of PCI and WCI (Lanson and Besson 1992) and since in our samples I-S peak influences (or overlaps) little on both PCI and WCI peaks (Fig. 5b), unlike the case of Lanson (1997). Here, we note that the higher LI and MLI indicate the higher illite crystallinity, reversely in KI.

On the one hand, the relative amount of smectite to illitic minerals or to chlorite was determined using peak intensities of the individual XRD peaks of smectite (Sm), PCI, WCI and chlorite (Ch) in EG patterns, and are expressed as Sm/(Sm+PCI+WCI) or Sm/(Sm+Ch), respectively. The relative amount of kaolinite (Ka) to illitic minerals or to chlorite was also determined using the individual peak intensities corresponding to their minerals, and are expressed as Ka/(Ka+PCI+WCI) or Ka/(Ka+Ch), respectively.

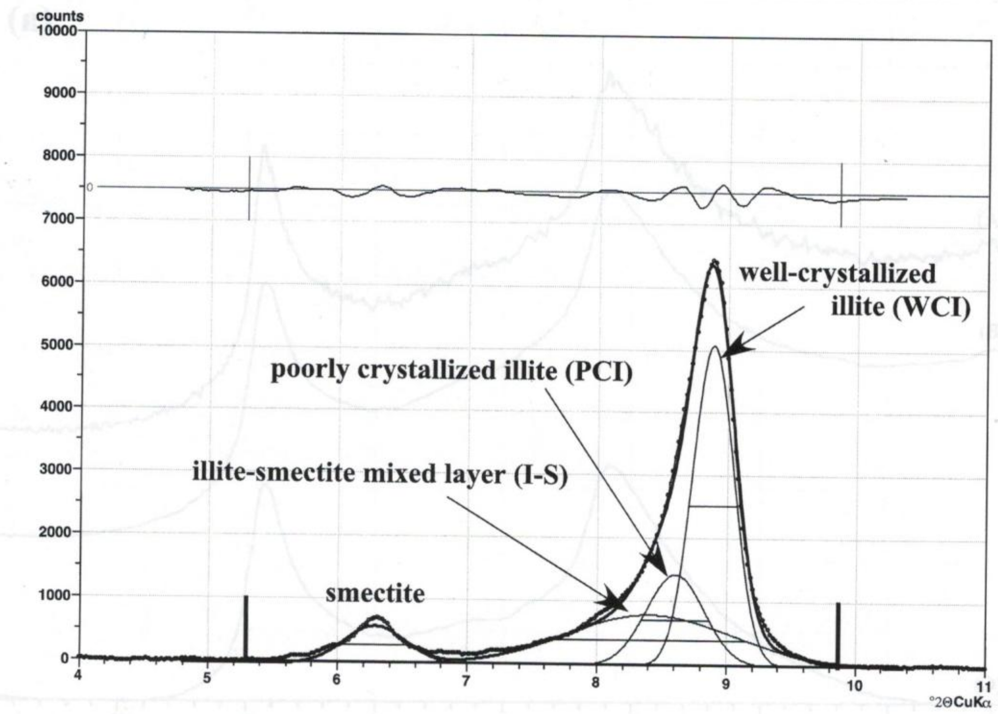

Fig. 4: An example of decomposition with four elemental peaks of XRD pattern in Fig. 1.

RESULTS AND DISCUSSION

Fig. 7 shows the variations of LI, MLI, Sm/(Sm+PCI+WCI), and Sm/(Sm+Ch) recorded in clay samples from 5 m to 49 m in depth of JW-3. The variations of the hydrolysis condition expected from LI and MLI are congruous with those of the hydrolysis condition expected from the relative amount of alteration minerals to primary minerals (Sm/(Sm+PCI+WCI) and Sm/(Sm+Ch)). In the weathering processes of the primary phyllosilicates (micas, chlorites), they are first exfoliated by hydrolysis, giving way to "exfoliated" illite and chlorite (Chamley 1989). If hydrolysis continues or becomes more active, the illitic minerals exhibit the degradation of the crystallinity and transform to illitic mixed-layer minerals (I-S, illite-vermiculite mixed layer, etc.) and finally to smectite. Hence, if the weathering of the phyllosilicates proceeds naturally, the variations of both illite crystallinity indices (LI and MLI) and the relative amount of alteration minerals to primary minerals (Sm/(Sm+PCI+WCI) and Sm/(Sm+Ch)) should link together. Therefore, it is strongly expected that these four indices accurately depict the hydrolysis condition recorded in the Kathmandu Basin sediments. The increasing hydrolysis conditions likely show wet climates, whereas the decreasing hydrolysis conditions probably correspond to drier climates.

On the other hand, the variations of the relative amount of kaolinite to illitic minerals or chlorite between 5 m and 49 m in depth of JW-3 show roughly mirror images of those of the relative amount of smectite to illitic minerals or chlorite, respectively (Fig. 8). Very similar results are reported in the study on clay minerals in the sediments of Lake Baikal (Yuretich et al. 1999). Yuretich et al. (1999) concluded that the clay facies characterized by relative increase of smectite and decrease of kaolinite indicates warm climates with vigorous chemical weathering and soil formation, whereas the clay facies showing relative decrease of smectite and increase of kaolinite represents colder climatic conditions, as deduced from the SPECMAP isotope curve. However, they have described that kaolinite does not necessarily argue against a cold climate, because kaolinite is a very resistant minerals and thus could be derived from physical weathering of kaolinite-bearing sedimentary rocks and erosion of palaeosols (Ehrmann et al. 1992). In addition, kaolinite is generally formed from feldspars which show larger particle size than clay size in the sediments (Chamley 1989). Therefore, we may need to see the relation between the variation of the amount of kaolinite and feldspar in the bulk samples, in order to ascertain whether kaolinite shows correlation with palaeoclimate in this region or not.

Fujii and Sakai (2001) showed that a palaeoclimatic variation based on pollen analysis of the sediments from JW-3 is congruous with a fluctuation pattern of δ^{18}O obtained from planktonic foraminifers of deep sea sediments in the Arabian Sea (Prell et al. 1992). They also estimated the sedimentation rate of the upper 115 m of JW-3 at about 104 mm/kyr. Then, we also attempted to compare a variation curve on the hydrolysis condition inferred from our four indicators (LI,MLI, Sm/(Sm+PCI+WCI), and Sm/(Sm+PCI+WCI)) with the variation curve of δ^{18}O, at the same time scale (Fig. 9).

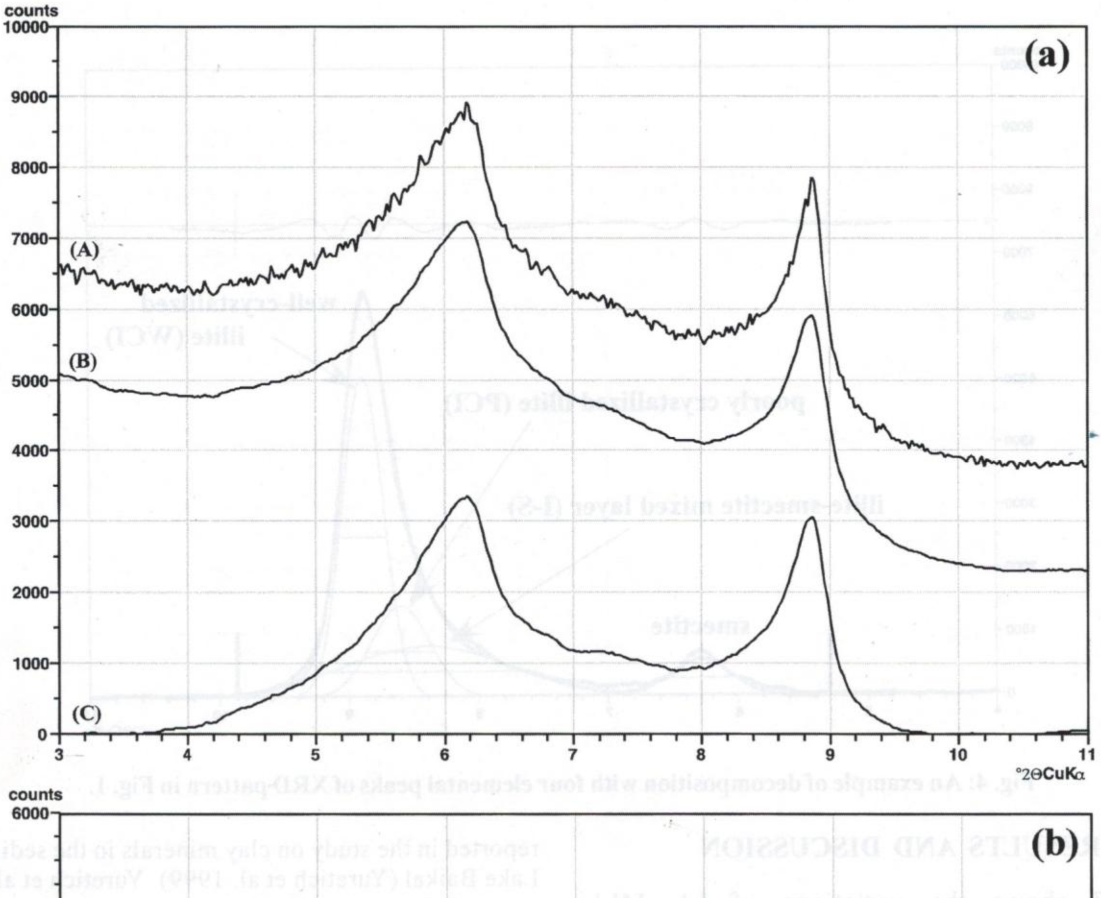

Fig. 5: Process of decomposition procedure for an XRD pattern of clay minerals in the drilled sediments (AD sample from 20 m depth sample). (a) (A): An XRD raw pattern, (B): XRD pattern after application of profile smoothing for (A), (C): XRD pattern after application of background stripping for (B). (b) A result of decomposition with five elementary peaks for the XRD pattern (C) in (a). I-S: illite-smectite mixed layer minerals, PCI: poorly crystallized illite, WCI: well-crystallized illite.

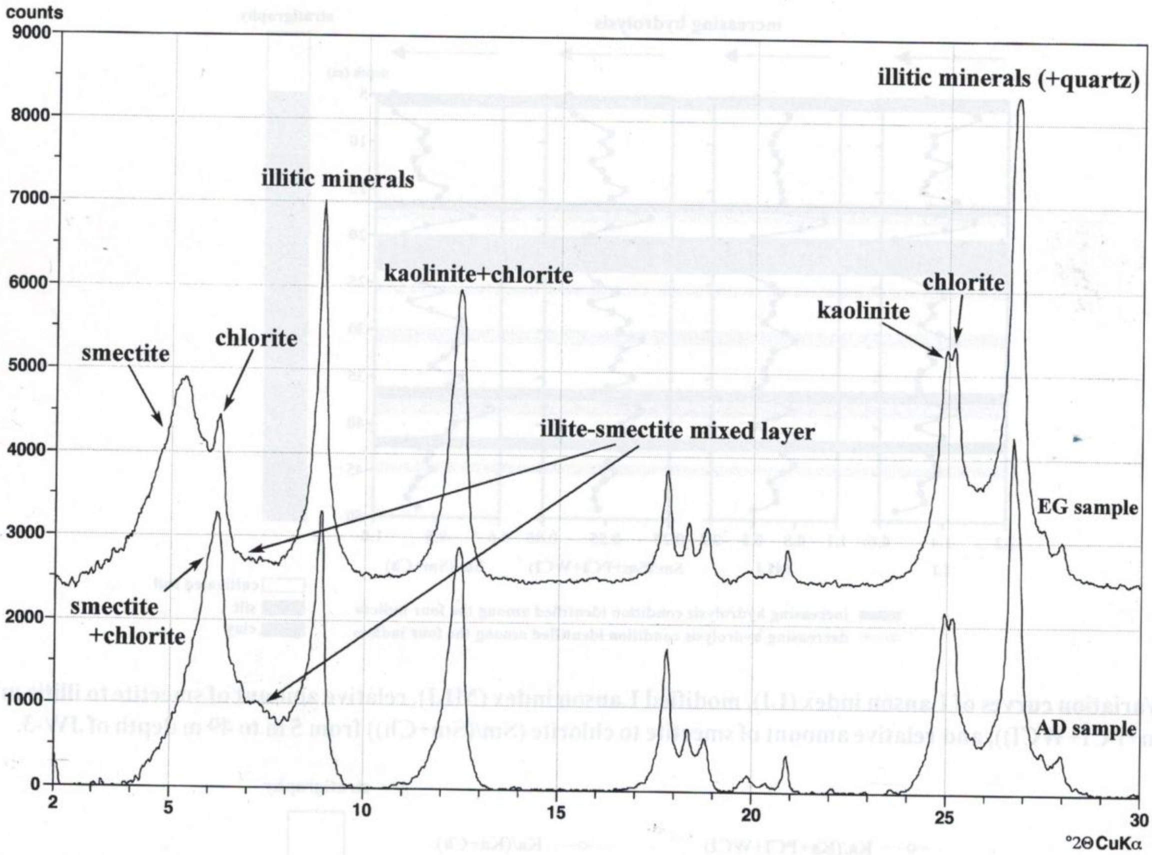

Fig. 6: Representative XRD patterns of AD and EG samples of clay minerals in the drilled sediments (20 m depth sample).

The variation of the hydrolysis condition is exceedingly in harmony with the variation of $\delta^{18}O$ from planktonic foraminifers of deep sea sediments in the Arabian Sea. The hydrolysis condition between 5 m and 49 m in depth of JW-3 shows five times of main fluctuation exhibiting increasing and decreasing one, with an interval of 8–12 m. The increasing hydrolysis conditions correspond to warm climates expected from the results of $\delta^{18}O$, while the decreasing hydrolysis conditions correspond to cold climates. These facts confirm that the climatic variations in the Kathmandu Basin were closely related to global climate, without apparent local climatic changes.

It was deduced from the oxygen isotope and pollen records from the Arabian Sea that a dry climate prevailed during the glacial period and a wet climate in the interglacial period (Van Campo et al. 1982). Also in the Kathmandu Basin, it is interpreted from pollen analysis that it was dry during the cold climate and wet in the warm climate (Fujii and Sakai 2001). In contrast, Niitsuma et al. (1991) proposed that climate in the Arabian Peninsula was more humid during the glacial period. As mentioned above, the increasing hydrolysis condition should have prevailed during wetter climate whereas the decreasing one should have been produced during drier climate (Chamley 1989). Our results clearly show that warm climates expected from the results of $\delta^{18}O$ correspond to increasing hydrolysis conditions while cold climates correspond to decreasing hydrolysis. Therefore, we conclude that it was dry (decreasing hydrolysis condition) during the cold climate and wet (increasing hydrolysis condition) during the warm climate in the Kathmandu Basin during the last about 450 kyr. Therefore, it is naturally expected that a cold-and-dry climate retarded hydrolysis (weathering) of minerals and a warm-and-wet climate intensified the hydrolysis in the Kathmandu Basin.

CONCLUSIONS

The present study shows that the decomposition procedure for XRD patterns of clay minerals is very useful for the accurate determination of the crystallinity and the relative amount of clay minerals. Our indices on the weathering or hydrolysis condition obtained by the decomposition procedure well reconstructed the palaeoclimatic variations recorded in the Kathmandu Basin sediments.

In the PKL project, we have carried out academic drilling within the Kathmandu Basin and have investigated on the cores and surface exposures from various viewpoints and methods (Sakai et al. 2000). If the decomposition procedure as well as other methods is performed for the drilled samples, the palaeoclimatic variations will be more accurately reconstructed.

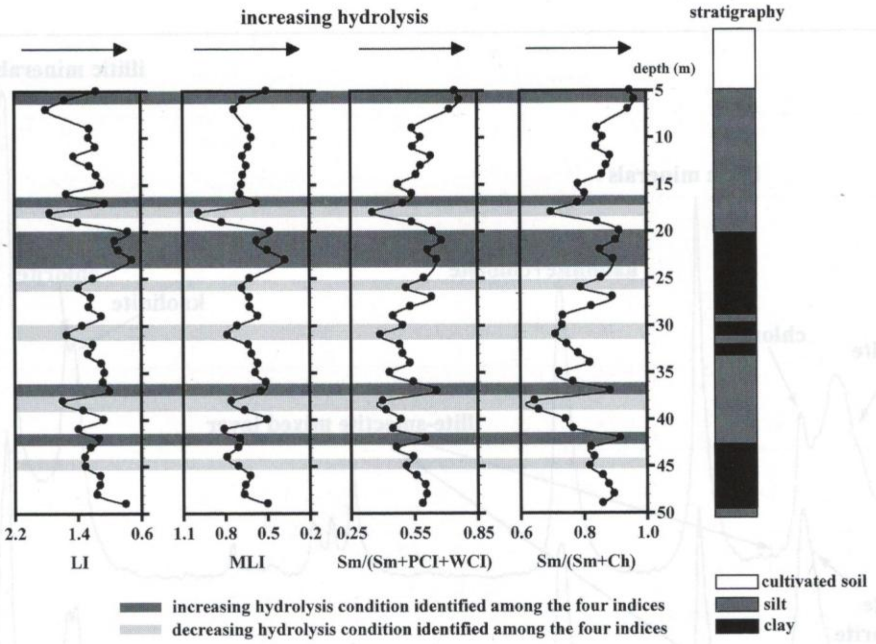

Fig. 7: Variation curves of Lanson index (LI), modified Lanson index (MLI), relative amount of smectite to illitic minerals (Sm/(Sm+PCI+WCI)), and relative amount of smectite to chlorite (Sm/(Sm+Ch)) from 5 m to 49 m depth of JW-3.

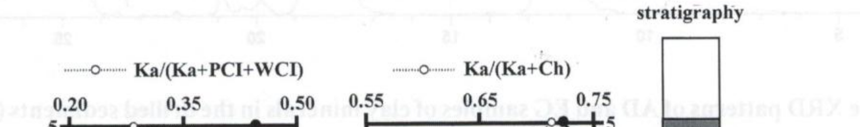

Fig. 8: (a) Comparison of variation curve of relative amount of smectite to illitic minerals (Sm/(Sm+PCI+WCI)) and that of relative amount of kaolinite to illitic minerals (Ka/(Ka+PCI+WCI)) from 5 m to 49 m depth of JW-3. (b) Comparison of variation curve of relative amount of smectite to chlorite (Sm/(Sm+Ch)) and that of relative amount of kaolinite to chlorite (Ka/(Ka+Ch)).

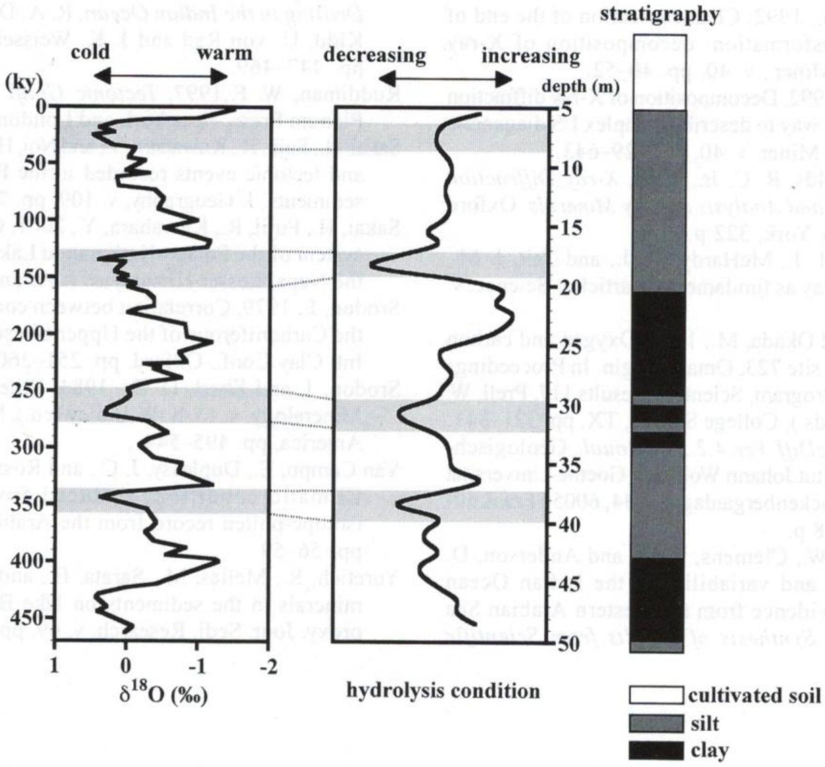

Fig. 9: Comparison of a variation curve of $\delta^{18}O$ from Ocean Drilling Program, site 722 (Prell et al. 1992) (a) and that of hydrolysis condition recorded in clay minerals from 5 m to 49 m in depth of JW-3, deduced by taking an average of our four indices (LI, MLI, Sm/(Sm+PCI+WCI), Sm/(Sm+Ch)) (b). The shaded regions in (a) and (b) correspond to colder climates and more decreasing hydrolysis conditions, respectively.

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