Quaternary Research 77 (2012) 368–381

Contents lists available at SciVerse ScienceDirect

Quaternary Research

journal homepage: www.elsevier.com/locate/yqres

Clay mineral variations in Holocene terrestrial sediments from the Indus Basin

Anwar Alizai [a,*], Stephen Hillier [b], Peter D. Clift [a], Liviu Giosan [c], Andrew Hurst [a], Sam VanLaningham [d], Mark Macklin [e]

[a] School of Geosciences, University of Aberdeen, Meston Building, Aberdeen, AB24 3UE, UK
[b] The James Hutton Institute, Craigiebuckler, Aberdeen AB15 8QH, UK
[c] Woods Hole Oceanographic Institution, Woods Hole, MA 02543, USA
[d] School of Fisheries and Ocean Sciences, University of Alaska Fairbanks, Fairbanks AK 99775-7220, USA
[e] Institute of Geography and Earth Sciences, University of Wales, Aberystwyth, UK

ARTICLE INFO

Article history:
Received 14 December 2010
Available online 23 February 2012

Keywords:
XRD
Clay mineralogy
Monsoon
Himalaya
Indus Delta
Floodplain
Fluvial processes
Large rivers

ABSTRACT

We employed X-ray diffraction methods to quantify clay mineral assemblages in the Indus Delta and flood plains since ~14 ka, spanning a period of strong climatic change. Assemblages are dominated by smectite and illite, with minor chlorite and kaolinite. Delta sediments integrate clays from across the basin and show increasing smectite input between 13 and 7.5 ka, indicating stronger chemical weathering as the summer monsoon intensified. Changes in clay mineralogy postdate changes in climate by 5–3 ka, reflecting the time needed for new clay minerals to form and be transported to the delta. Samples from the flood plains in Punjab show evidence for increased chemical weathering towards the top of the sections (6–<4 ka), counter to the trend in the delta, at a time of monsoon weakening. Clay mineral assemblages within sandy floodplain sediment have higher smectite/(illite + chlorite) values than interbedded mudstones, suggestive of either stronger weathering or more sediment reworking since the Mid Holocene. We show that marine records are not always good proxies for weathering across the entire flood plain. Nonetheless, the delta record likely represents the most reliable record of basin-wide weathering response to climate change.

Introduction

Variations in climate are one of the most prominent drivers of change in continental environments. Reconstructing the response of landscape to climate change over long periods of geological time is often difficult because of fragmentary sedimentary records, and because of the paucity or even complete lack of age control in terrestrial sediments. As a result, marine sediments are often used to constrain evolving continental environments, based on the assumption that the sediments that accumulate on a continental margin at any given time are representative of the terrestrial drainage basins from which they are sourced and the processes occurring therein (Thiry, 2000). However, if the flux of sediment at the delta largely reflects reworking of older flood-plain deposits, or if the sediment transport times are long and punctuated, then this assumption may not be valid. In such circumstances marine sediments would not act as good proxies of continental weathering conditions.

In this study we assess the linkage between marine and terrestrial clay mineral records in a single major river system. To do this we present clay mineral data from across the flood plains and contemporaneous deltaic sediments from the Indus River basin in SW Asia (Fig. 1). We chart how clay mineral assemblages have changed onshore in response to evolving monsoon intensity since ~14 ka. We further compare those mineralogies deposited in the flood plains with those at the delta in order to establish if the marine record evolves coherently with the flood plain, or not. By studying both the potential source areas and the delta sink, we establish a better understanding of the constraints and the caveats on the use of clay mineral assemblages as proxies of continental environmental change, a perspective that is not available to most studies of marine sediments that are typically made without the onshore controls enjoyed here.

The purpose of the current study is to document temporal variations in the clay mineralogy of the terrestrial sediments of the Indus system during the Holocene and to determine how these might be linked to previously published reconstructions of climatic variations since the end of the Last Glacial Maximum (LGM: ~20 ka) (Overpeck et al., 1996; Fleitmann et al., 2003; Gupta et al., 2003). Here we report for the first time on how monsoon variations since 14 ka correlate with changing clay mineral compositions in a major river flood plain, feeding one of the world's largest delta systems. In so doing we provide a linked continental source to marine sink perspective on the interpretation of clay mineral records in terms of environmental evolution, a perspective that is rarely considered, or indeed available when older marine sequences are analyzed.

* Corresponding author.
E-mail address: anwar.alizai@gmail.com (A. Alizai).

0033-5894/$ – see front matter © 2012 University of Washington. Published by Elsevier Inc. All rights reserved.
doi:10.1016/j.yqres.2012.01.008

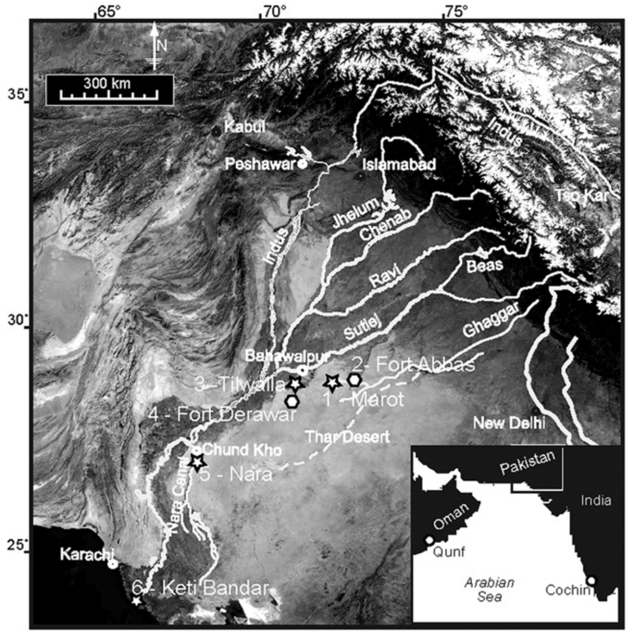

Figure 1. Study area showing four core sites (stars) and two trenches (hexagons) in the Indus River floodplain. Inset map provides regional overview, as well as the location of sites mentioned in the text outside the Indus basin.

Clay mineral weathering proxies

There is general consensus among clay mineralogists that the neo-formation and transformation of clay minerals in soil profiles and regoliths is determined by an interaction between the geology, geomorphology (insofar as it controls drainage) and the climate of the source terrain (Morgan, 1973; Singer, 1984; Hillier, 1995; Wilson, 1999). Thus global-scale maps of mineral distribution on the deep-sea floor show that the general zonation of clay minerals mirrors contemporary climates on the continents (Biscaye, 1965; Griffin et al., 1968; Hamann et al., 2009). This implies that climatic conditions are a primary control on clay mineralogy, and that temporal changes in climate have the potential to cause changes in the clay mineralogy of the clastic component of the run-off (Singer, 1984; Chamley, 1989; Thiry, 2000). An outstanding question is how fast the response time of the weathering is to climate change and if marine clay mineralogy can record changes on millennial scales.

Before using clay mineral assemblages to interpret paleoclimate, however, a number of assumptions have to be made. One is that clay mineral formation is a direct response to climatic conditions (Thiry, 2000) and the time required to respond to changes in climate is shorter than the time interval being examined. This assumption may become less valid as shorter time periods are considered (<1000 yr), simply because the formation of new clays, or the transformation of existing clay minerals, are both dependent on weathering reactions with finite rate-determining steps (Fig. 2). Sediment-transport and residence times also may be an additional source of uncertainty, although this is likely to be less important on time scales $>10^6$ yr. Clay mineral records from the South China Sea show a good correspondence between clay assemblages and the intensity of the East Asian monsoon over periods $>10^6$ yr (Wan et al., 2007). Recently, it has also been suggested that changes in mineralogy linked to millennial-scale variations are recorded in sediments from other Asian continental margins (Boulay et al., 2007; Colin et al., 2010), implying that the clay minerals in a weathering system can respond and leave a record on these shorter time scales too. We further assume that inherited clay minerals give information about the environment from which they have been derived (Hillier, 1995) and that, once formed, clay minerals remain stable unless altered by diagenetic processes, i.e. post-sedimentary processes.

This study was designed to examine how clay mineralogy within the Indus basin might reflect the evolving Holocene climate, principally the intensity of the SW Asian summer monsoon. Does weathering respond rapidly to postglacial climate change? We use illite crystallinity to trace the intensity of weathering because this is commonly interpreted as an index of the hydrolyzing power of the (soil) environment from which the mineral is derived (Lamy et al., 1998). Higher temperatures and rainfall leads to leaching by strong hydrolyzation, resulting in low crystallinity (wider XRD peaks). Thus, relative changes in the crystallinity of detrital illite can potentially help to differentiate between more or less weathering and thus to cold-dry and warm-humid conditions. We employ this method to assess whether monsoon intensity affects weathering over time scales of ~1000 yr in the manner anticipated from longer-term studies, because a strong monsoon might be expected to favor strong hydrolyzation.

In addition to illite crystallinity, we ratio a variety of clays whose origins are believed to be different and which have been used in the past as indicators of weathering intensity. Illite and chlorite are generally considered to be the products of physical erosion of low-grade metamorphic rocks with little chemical weathering and alteration. Illite distributions in marine sediments are related to detrital rather than authigenic processes (Griffin et al., 1968; Rateev and Gorbunova, 1969). Similarly chlorite, frequently associated with illite, is notably more abundant in present-day high-latitude soils and sediments (Biscaye, 1965; Jacobs, 1970). A clay–mineral assemblage rich in chlorite and illite is therefore typical of soils and sediments produced in high latitudes or by cold-climate weathering because chemical weathering in such settings is weak (Bockheim, 1982; Campbell and Claridge, 1982).

In contrast, soil forms rapidly and to greater depths in tropical and subtropical environments, where chemical weathering is intensified by the process of leaching (Birkeland, 1984). As a result, kaolin-group minerals and gibbsite are frequently abundant in well-developed (meters thick) soils from regions of tropical climate with high rainfall, whereas warm, dry regions with less leaching dominantly produce smectite-rich soil. Chlorite and illite prevail at high latitudes where physical weathering dominates (Thiry, 2000). Soil formation in arid areas, both cold and warm, is insensitive to climatic conditions; hence, paleoclimate reconstruction in these regions is generally not feasible using soils (Thiry, 2000). The kaolinite/chlorite ratio in marine sediments constitutes a reliable indicator of chemical hydrolysis versus physical processes in continental weathering profiles (Chamley, 1989). Kaolinite/chlorite and kaolinite/illite are proxies of humidity and high values are indicative of enhanced humidity (Thamban et al., 2002; Thamban and Rao, 2005).

When a soil is eroded its clays may be transported to the continental margin. This explains why in recent deep-sea sediments of the North and South Atlantic the distribution of physically eroded chlorite has been observed to be nearly reciprocal to that of neo-formed, soil-derived kaolinite (Biscaye, 1965; Zimmermann, 1977). These patterns in the world oceans represent the signals of clay mineral change caused by large differences in climate. Over smaller regions, or with less extreme climate variation, the sedimentary record of weathering is likely to be manifested by more subtle changes in clay mineralogy. For example, changes in the relative abundances of clay minerals along a climatic gradient (280–720 mm average annual modern rainfall, mean annual temperature 9–13°C) across the Chinese Loess Plateau, related to variations in the Asian Monsoon, showed kaolinite increasing from around 2–3 to 5–6%, while expandable (smectitic) clays increased from ~30 to 60%, chlorite decreased from 7–8 to 0–3%, and illite (micas) decreased from 40–50% to 20–30% (Jeong et al., 2011). It is not clear if less dramatic changes would register as a clear paleoclimatic signal in sediments because

https://doi.org/10.1016/j.yqres.2012.01.008 Published online by Cambridge University Press

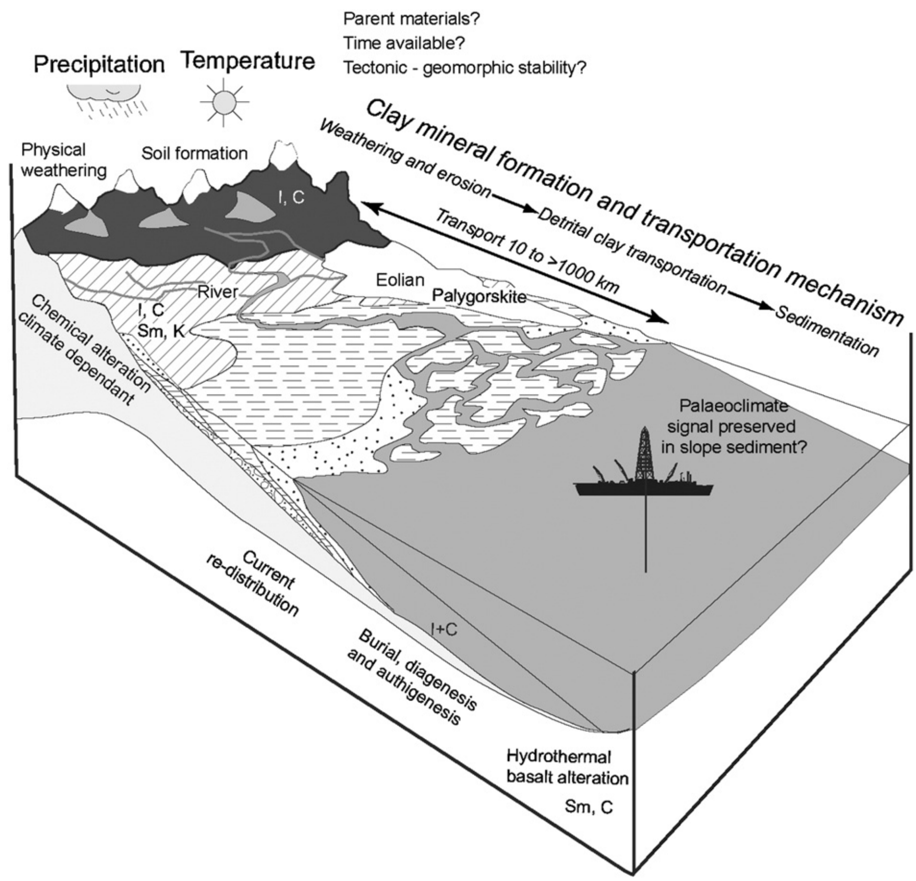

Figure 2. Clay–mineral formation and transport mechanism modified after Fagel (2007). This cartoon shows the many processes and factors that must be considered when trying to extract paleoclimatic information from clay minerals, which may be linked directly or indirectly to climatically modulated surface processes. I (illite), C (chlorite), Sm (smectite) and K (kaolinite) may also reveal information about the source.

the normal but complex processes of weathering must combine with episodes of erosion, mixing, storage, and transport before ultimate deposition.

The Indus River basin

The Indus Basin is a good location for a study of the clay minerals in both the flood plains and delta of a major river because it lies under the influence of the SW monsoon and experiences large volumes of sediment transport from the western Himalaya and Karakoram, which are experiencing rapid erosion. Although the region experiences strong summer rain it is relatively arid compared to the Ganges–Brahmaputra basin further east (Bookhagen and Burbank, 2006). Samples were taken from four core sites and two shallow trenches in the Indus flood plains that have been the focus of earlier provenance and sedimentology studies (Alizai et al., 2011b; Clift et al., in press). The core sites, each about 50 m deep, and the two shallow trench sites each ~4 m deep, are located in the upper eastern Pakistani Punjab, near the city of Bahawalpur (Fig. 1). Sands >5 m thick are interpreted as channel-fill deposits, while interbedded shales and thin sands correspond to overbank deposits (Fig. S1, Online data supplement). Because channels meander and migrate through time the appearance of sand in a single section does not necessarily indicate a basin-wide increase in sediment flux from the mountains.

The sites lie southeast of the modern course of the Sutlej River on the edge of the Thar Desert, but are located in the paleo-flow path of the Ghaggar-Hakra River, which is now an ephemeral course that may have previously contained a year-round flow (Ghose et al., 1979; Valdiya, 2002). The sample sites in the Punjab were designed to provide a weathering record of conditions in the eastern flood plain during the Holocene close to where the Ghaggar-Hakra may have flowed. This location on the edge of the Thar Desert means that conditions here are especially sensitive to changes in summer monsoon strength.

As well as the Punjabi sites we analyzed materials from a third site located further south, 13 km south of the village of Chund Kho in the region of the Nara Canal (Fig. 1). At this site, coring recovered material to a depth of 30 m. The Nara region lies just south of the modern confluence of the major eastern and western tributaries of the Indus, prior to its arrival at the delta in the Arabian Sea, however, the Nara Valley is no longer an active strand of the modern river and its geometry is reminiscent of capture from a separate, earlier, drainage. As a result, the sediments cored in the Nara Valley may represent the flood-plain deposits of an earlier river, undoubtedly of Himalayan provenance, but not necessarily the main Indus stream (Alizai et al., 2011b).

The southernmost site considered is located in the Indus delta at Keti Bandar, close to the mouth of the Indus River at the Arabian Sea coast (Fig. 1). Although the site is no longer a location of active accumulation, previously published ^{14}C dating has established that sedimentation was continuous from ~13,500 to 300 'calendar' years before present (cal yr BP) (Clift et al., 2008) (Online data supplement; Figs. S1 and S2). Because the Indus delta has prograded seawards since ~13.5 ka (Giosan et al., 2006) changes in sedimentation rates and in lithology are mostly linked to shoreline advance and retreat rather than variations in the rate of sediment supply.

Thus, these three regions provide three localized areas where we can examine the clay mineralogy of the sediment being transported and stored within the Indus River basin during the Holocene. We

https://doi.org/10.1016/j.yqres.2012.01.008 Published online by Cambridge University Press

test whether the clay mineral changes at the river mouth are similar to those preserved synchronously around the Thar Desert and determine if these can be linked to changes in climate. In so doing we consider the degree of sediment buffering in the flood plains and integration by the delta. Geomorphic studies have highlighted the fact that the flood plains have experienced incision and recycling during the Holocene (Clift et al., 2009), which may have influenced the mineralogy of the sediment reaching the ocean since these sediment would be expected to be more weathered than fresh material from mountain sources.

In order to interpret trends in clay mineralogy in terms of weathering changes we must first understand the potential effects of changing sources. Earlier provenance work from the flood plains is based on U–Pb dating of zircon grains (Alizai et al., 2011a; Clift et al., in press) and the Pb isotope characteristics of K-feldspar grains (Alizai et al., 2011b). However, these studies refer only to sands, not the clays targeted here. The difference is significant because sand, especially dense grains like zircon, tends to be transported as bedload rather than as suspended sediment, as is the case with clays. These earlier studies show a general pattern of sediment supply from the Himalaya-draining Yamuna and Sutlej Rivers to the eastern parts of the flood plains (Marot; Fig. 1) and from the similarly sourced Beas River to regions just to the west (Tilwalla). Both these boreholes show increasing flux from the Thar Desert going up-section prior to the deposition of the last channel sands. Sediments at Fort Abbas are believed to largely represent reworking from the Thar Desert (Clift et al., in press).

Only at Keti Bandar is there Nd isotope data from finer-grained materials to help constrain provenance. This is important because at Keti Bandar sediment flux is integrated from across the whole basin. The study of Clift et al. (2008) showed a shift away from Karakoram-derived sediment towards sources in the Himalaya between the base of the core and ~50 m depth (~7–9 ka) above which level they demonstrated relative stability from that time to the present day. Thus changes in clay mineralogy at Keti Bandar above 50 m depth can be most clearly linked to changes in environmental conditions rather than provenance.

Methods

X-ray diffraction (XRD) is the most reliable and commonly used technique for the determination and quantification of clay mineral assemblages. In this study we use the semi-quantitative method of Moore and Reynolds (1989) to estimate the clay assemblage, which is based on peak-intensity factors determined from calculated XRD patterns. Hillier (2003) described the method used in the present study in detail, including its validation and an assessment of analytical uncertainty based on its application to prepared mixtures of pure clay minerals. For clay minerals present in amounts >10 wt.% uncertainty is estimated as better than ±5 wt.% at the 95% confidence level. Uncertainty of peak area measurement based on repeated measurements is typically <5%, with the smallest peaks having the highest uncertainties. In addition to calculating the relative percentages of clay minerals in the samples, we also determine various ratios based on measurements of peak areas in the XRD patterns. These ratios are used to reconstruct changes in relative abundance and are directly proportional to ratios calculated from the exact values for individual mineral percentages. Data are presented as both relative percentages and as ratios to best illustrate factors that control trends.

184 samples were taken from four core sites and two trenches. These comprise drill sites at Marot (44 samples), Tilwalla (63 samples), Nara (14 samples) and Keti Bandar (30 samples), as well as trench sites at Fort Abbas (19 samples) and Fort Derawar (14 samples) (Fig. 1). The clay-sized fraction (<2 μm) was separated by sedimentation and application of Stokes' Law. The filter-transfer method was used to orient clay minerals on a glass slide prior to X-ray analysis following the method of Moore and Reynolds (1989). The samples were analyzed at the Macaulay Institute, Aberdeen, UK (now the Hutton Institute), using a Siemens X-ray diffractometer, with Co Kα radiation selected by a diffracted beam monochromator. A set of three diffraction patterns (i.e., air-dried, glycolated and heated to 300°C for 1 h) were produced for each sample and used for mineral quantification. All patterns were collected by step scanning from 2 to 45° 2θ, in 0.02° steps and counting for 1 s per step. Peak area measurements for individual minerals were made using Bruker Diffrac Plus EVA-12.0 software. Ratios reflecting relative variations in clay mineral abundance were determined from peak area measurements. Illite crystallinity was measured using the Bruker Eva software as the full width at half maximum (FWHM) of the illite 001 basal peak in glycolated XRD patterns in units of Δ°2θ; formally known as the 'Kubler' index. We also measured the integral breadth of the illite 001 peak, which is the breadth in Δ°2θ of a rectangle of the same area and height as the corresponding peak. Integral breadth trended in the same fashion as FWHM and is not discussed further in this paper.

Depositional age determinations were taken from previous studies and were mostly made by the ^{14}C dating method to mollusc shells, as well as to wood and plant remains. These were further supplemented by optically stimulated luminescence dating. The compilation is given in Table 1 and shown graphically in the online data supplement (Fig. S1). Depositional ages for the Keti Bandar core site were

Table 1
Radiocarbon ages from the core and trench sites used to control the timing of sedimentation in the studied sections. Ages at Keti Bandar are from Clift et al. (2008), while those at Tilwalla are from Clift et al. (in press). The ages from Marot, Nara and Fort Abbas are from Alizai et al. (2011b).

Sr. no.	Location	Sample ID	Material type	Depth (m)	^{14}C ages (^{14}C yr BP)		Calendar age range (2 sigma, cal yr BP)
1	Keti Bandar	KB-6-2	Plant/wood	15.48	245	±25	277–317
2	Keti Bandar	KB-11-1	Mollusc shell	29.62	2560	±30	1813–1947
3	Keti Bandar	KB-17-2	Mollusc shell	49.55	7080	±40	7153–7427
	Keti Bandar	KB_19-4	Mollusc shell	55.49	8210	±55	9017–9309
4	Keti Bandar	KB_19-4	Mollusc shell	55.49	8710	±65	8302–9139
5	Keti Bandar	KB-23-2	Mollusc shell	67.24	9160	±35	8850–9687
6	Keti Bandar	KB-28-2	Mollusc shell	80.33	10,100	±30	10,477–10,772
7	Keti Bandar	KB-35-1	Mollusc shell	100.22	12,650	±40	12,953–13,981
8	Keti Bandar	KB-38-3	Mollusc shell	110.73	13,000	±55	13,265–15,172
9	Keti Bandar	KB-40-4	Mollusc shell	116.93	28,700	±110	31,623–30,711
10	Keti Bandar	KB-40-4	Coral	116.93	38,900	±410	42,032–40,572
11	Fort Abbas	080421-3	Mollusc shell	3.48	5050	±35	3957–3767
12	Nara	NA-7-7	Bivalve	6.	4830	±25	3660–3531
13	Marot	MAR-2-5	Gastropod	9.37	6500	±50	5604–5359
14	Tilwalla	Till-3-17B	Plant/wood	43.7	45,100	±670	47,980–44,801

All the radiocarbon data were produced at the National Ocean Sciences Accelerator Mass Spectrometry Facility (NOSAMS) at the Woods Hole Oceanographic Institution, USA. Keti Bandar ages after Clift et al. (2008).

https://doi.org/10.1016/j.yqres.2012.01.008 Published online by Cambridge University Press

published by Clift et al. (2008); ages from Marot, Tilwalla, Fort Abbas and Nara were published by Clift et al. (in press) and Alizai et al. (2011b).

Results

The major clay mineral groups resolved at all study sites are illite, kaolinite chlorite and smectite. The identification of illite, chlorite and kaolinite was straightforward, whereas for smectite there was some variation in the diffraction patterns that may indicate a small degree of mixed layering in some samples. Typically, this is manifest by subtle changes in peak positions and peak profiles that appear to vary in relation to the relative abundance of smectite in a given sample. Thus samples characterized by a high relative abundance of smectite show rational peak positions that are consistent with a pure 'end-member' smectite (Moore and Reynolds, 1989). There is no single peak position for end-member smectite, the key to its precise identification are the rational peak positions (i.e., the 005 peak is an exact submultiple of the 001 spacing). In contrast, samples with relatively low smectite abundance show peak positions with a measureable degree of irrationality (irregular peak position) and correlative changes in peak shape. These characteristics indicate unequivocally that the clay mineral is not pure (end-member) smectite but contains a small degree of (<20%) of mixed layering, probably of illite layers. However, for the sake of simplicity, both types are referred to as 'smectite' and the evidence for mixed layering is referred to specifically when relevant. The clay mineral data is divided into three geographical areas, which we present independently before discussing them together.

The general character of the clay assemblages is shown in Figure 3, which demonstrate the generally low levels of kaolinite seen in all samples, but the wide range in smectite compared to illite and chlorite. The samples from Keti Bandar have a more restricted range than many of the flood-plain sites. The highest proportions of smectite are found at Nara, Tilwalla and Marot, although all three sites also have examples of some of the most (illite + chlorite)-rich assemblages.

Mineralogy in the upper eastern Punjab

The stratigraphy sampled at the two drilling sites south of Bahawalpur show that they both have similar clay mineral assemblages.

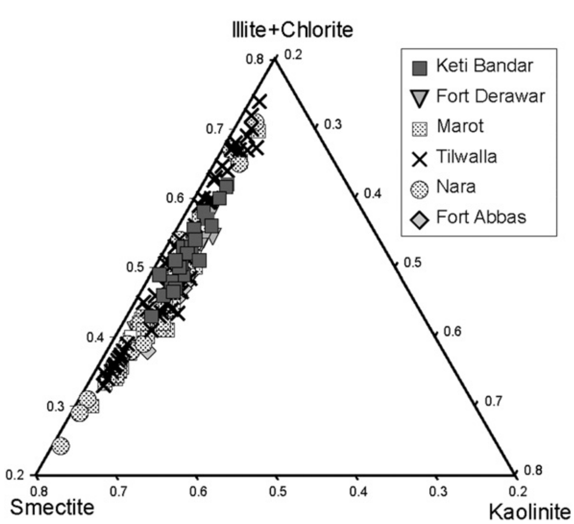

Figure 3. Ternary diagram of clay–mineral compositions from all the samples considered in this study. They show uniformly low proportions of kaolinite and a wide range in smectite versus physically weathered illite and chlorite clays. Note that clay–mineral variation observed at Keti Bandar is less extreme than that found in the flood plains in Punjab.

Clay mineral weight percentages at Marot comprise smectite (27–64%), illite (25–60%), kaolinite (2–6%) and chlorite (4–11%) (Table S1). At the Tilwalla core site the clay mineral assemblage comprises smectite (25–65%), illite (24–66%), kaolinite (1–6%) and chlorite (5–12%). The trench at Fort Abbas shows clay mineral abundances of smectite (47–57%), illite (32–45%), kaolinite (3–5%) and chlorite (6–8%). The other trench site at Fort Derawar comprises smectite (39–57%), illite (38–50%), kaolinite (2–5%) and chlorite (7–11%). Because they are the most abundant clay minerals present, most of the variations in relative abundance are caused by changes in the proportions of smectite and illite. As mentioned above, sediments with relatively low smectite abundance appear to be characterized by the presence of mixed-layer illite–smectite, whilst those with high smectite content are characterized by a more nearly pure end-member smectite. Two samples from the Tilwalla section that are illustrative of these types are shown in the online data supplement (Fig. S3).

The more subtle variations in the relative abundance of chlorite and kaolinite are more easily appreciated by an examination of peak area ratios. Clay ratios based simply on the areas of key peaks are useful in defining and comparing possible environmental trends. We have employed kaolinite/smectite, kaolinite/illite, smectite/illite, smectite/(illite + chlorite), kaolinite/chlorite and kaolinite/(illite + chlorite) ratios because these ratios are all potentially environmentally controlled and have been successfully used in past studies. Here we try to assess which of these is the most sensitive.

The evolving mineral ratios in the two cored sections allow these to be divided into three broad intervals. The upper section (0–14.16 m) in the Marot core, which is dominated by the finest grained sediments (Fig. 4), has kaolinite/smectite values of 0.06–0.19 (averaging 0.12). The second depth interval of 14.16–28.94 m comprises both fine and sand–sized material with kaolinite/smectite values ranging 0.05–0.17 (averaging 0.09). The deepest depth interval of 28.94–41.14 m has kaolinite/smectite values of 0.06–0.08 (averaging 0.08). These values are close to those found in the Tilwalla core over depth intervals 0–22.23 m, 22.23–37.48 and 37.48–45.11 m, where kaolinite/smectite ratios are 0.03–0.17, (averaging 0.10), 0.04–0.10 (averaging 0.06) and 0.05–0.11 (averaging 0.07) (Figs. 4B and 5B). Variation in kaolinite/smectite with depth at these two sites is also mirrored in the variability in illite crystallinity (FWHM) (Figs. 4A and 5A). Both sites show similar patterns in depth-lithology and depth-clay variations. It is noteworthy that kaolinite/smectite and kaolinite/illite ratios are generally higher at the top of both sections compared to deeper and they appear to be less affected by changes in lithology compared to some other ratios. In all the measured sections there is a clear correlation between clay mineralogy and lithology, most clearly with smectite being enriched and illite reduced over sandier intervals.

At Fort Abbas and Fort Derawar, the kaolinite/smectite ratio ranges 0.10–0.19 (averaging 0.15) and 0.07–0.19 (averaging 0.13) respectively, most similar to the shallowest part of the sections at Marot and Tilwalla. Other mineral ratios at Fort Abbas include kaolinite/illite of 0.12–0.26 (averaging 0.20), smectite/illite of 1.02–1.59 (averaging 1.34), kaolinite/(illite + chlorite) of 0.11–0.22 (averaging 0.17) and smectite/(illite + chlorite) of 0.9–1.48 (averaging 1.14) (Online Data Supplement: Fig. S4B). At Fort Derawar kaolinite/smectite is higher still compared to the sediments from Marot and Tilwalla, showing values of 0.07–0.19 (averaging 0.13). Kaolinite/illite values range 0.10–0.21 (averaging 0.14); smectite/illite ranges 0.79–1.66 (averaging 1.12), kaolinite/(illite + chlorite) is 0.08–0.16 (averaging 0.12) and smectite/(illite + chlorite) is calculated at 0.68–1.39 (averaging 0.94) (Online Data Supplement: Fig. S5B). These latter ratios are a little lower than those found at Fort Abbas.

Mineralogy in the Nara region

Clay mineral analysis of the Nara core (Online data supplement; Table S1) show smectite abundances at 27–74%, illite at 20–58%,

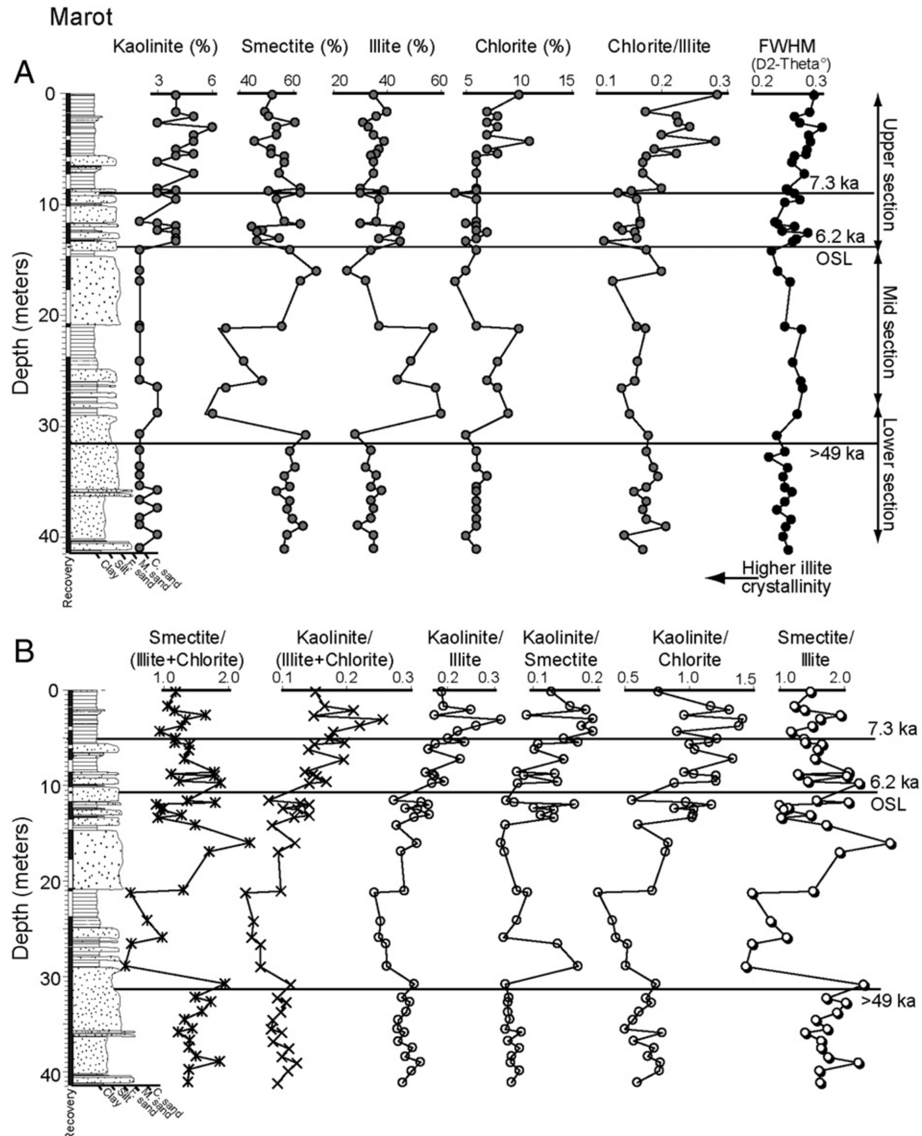

Figure 4. Down-core variation in clay minerals in the Marot borehole showing variations in the relative abundance of kaolinite, smectite, illite and chlorite along with chlorite/illite ratio and the crystallinity index for illite, as measured by the full width at half maximum (FWHM) of the illite 001 basal peak. (B) Variations in climate-sensitive proxy ratios: smectite/(illite + chlorite), kaolinite/(illite + chlorite), kaolinite/illite, kaolinite/smectite, kaolinite/chlorite and smectite/illite. Note increasing values of kaolinite/illite, kaolinite/smectite, and kaolinite/chlorite above 15 m depth.

kaolinite at 1–4% and chlorite at 4–13%. This is in the same general range as that seen in the Bahawalpur region, i.e. high smectite and illite and low kaolinite and chlorite. However, in the Nara area we see long amplitude variations in smectite and illite compared to the Bahawalpur area, while kaolinite and chlorite show more or less the same composition as further north.

At Nara the kaolinite/smectite, smectite/illite, kaolinite/illite, kaolinite/(illite + chlorite) and smectite/(illite + chlorite) ratios show coherent variability with depth, and a generally good match with changes in the illite crystallinity index. (Figs. 6A, B). The clay ratios allow us to divide the section into three intervals (i.e., 0–8.24, 8.24–23.21 and 23.21–29.39 m). In the upper interval crystallinity is low, as is smectite/illite and smectite/(illite + chlorite). Ratios involving kaolinite are generally more variable, likely because of the low concentrations of that mineral. Illite crystallinity, smectite/illite and smectite/(illite + chlorite) all increase below 8 m depth (close to the ^{14}C age of 3400–3700 cal yr BP). These ratios decrease further down-section, except for another high point close to 24 m depth and at the lowest sample at 29 m depth.

Mineralogy in the delta region

The section at Keti Bandar, representing flux to the delta, shows clay mineral abundances ranging from 35–54% for smectite, 34–52% for illite, 1–5% for kaolinite and 7–12% for chlorite, i.e. similar ranges to those found on the flood plains of the Punjab, indicating that the assemblages are essentially coherent from both areas (Online data supplement; Table S2). Illite increases in parallel with a concomitant decrease in smectite with depth. Smectite abundance is greatest and illite abundance is the least at ~48 m depth, which is just above the ~8200 cal yr BP flooding surface that separates the section here into two coarsening-upwards regressive cycles.

The average mineral ratios at Keti Bandar are kaolinite/smectite 0.11, smectite/illite 1.06, kaolinite/illite 0.12, kaolinite/(illite + chlorite) 0.10 and smectite/(illite + chlorite) 0.88. The average value for the illite crystallinity index (FWHM) is 0.31 (Fig. 7). Coherent variability with depth is seen for some of the ratios. Smectite/illite and smectite/(illite + chlorite) generally show low values towards the base of the core, except for the lowermost samples dated as having been exposed

https://doi.org/10.1016/j.yqres.2012.01.008 Published online by Cambridge University Press

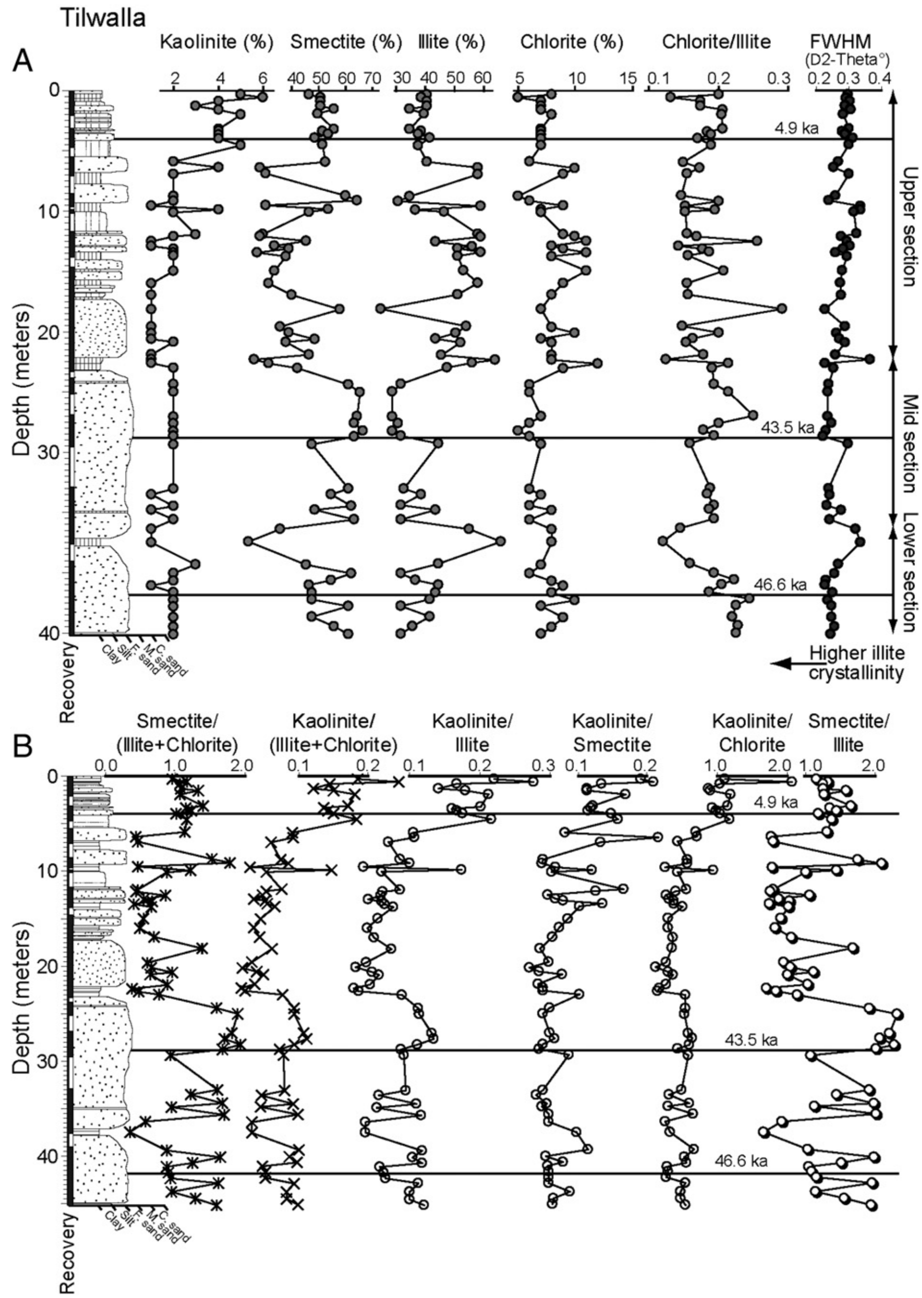

Figure 5. Down-core variation in clay minerals at the Tilwalla core site showing variations in the relative abundance of kaolinite, smectite, illite and chlorite percentages along with chlorite/illite ratio and the crystallinity index for illite. (B) Variations in climate-sensitive proxy ratios: smectite/(illite + chlorite), kaolinite/(illite + chlorite), kaolinite/illite, kaolinite/smectite, kaolinite/chlorite and smectite/illite.

and possibly deposited at the LGM (>20 ka), which lie below an unconformity separating them from the 14 ka overlying transgressive muds (Clift et al., 2008). These ratios show a gradual increase up-section to ~48 m depth, before falling from that point to the surface. Kaolinite/illite also shows similar trends, as does kaolinite/smectite (Fig. 7B), both of which fall sharply in the upper 30 m of the sediment column. This reflects the decrease in kaolinite abundance over the same depth interval.

Discussion

Regional variations in clay mineralogy can be assessed by plotting clay mineral ratios against one another. Figure 8A shows the variations in kaolinite/(illite + chlorite) compared to those in illite crystallinity for all the different study areas. This diagram shows that there is no overall close relationship between proxies for leaching and for alteration by hydrolization during weathering. All sites show a wide array of illite crystallinity, but there are resolvable differences between sites in kaolinite/(illite + chlorite). Sediments at Fort Abbas and Marot have especially high kaolinite/(illite + chlorite) values. Although sediments from Tilwalla also have high values it is noteworthy that the lowest kaolinite/(illite + chlorite) values are found there and in the Nara valley. The delta sediments from Keti Bandar cluster with moderate values in kaolinite/(illite + chlorite), suggesting that they integrate material from across the entire drainage, eliminating extreme variations that may occur locally. The differences among Tilwalla and the Marot and Fort Abbas samples may in part reflect their different source origins (Clift et al., in press), rather than radical differences among sites that are close by and experience similar climatic conditions.

https://doi.org/10.1016/j.yqres.2012.01.008 Published online by Cambridge University Press

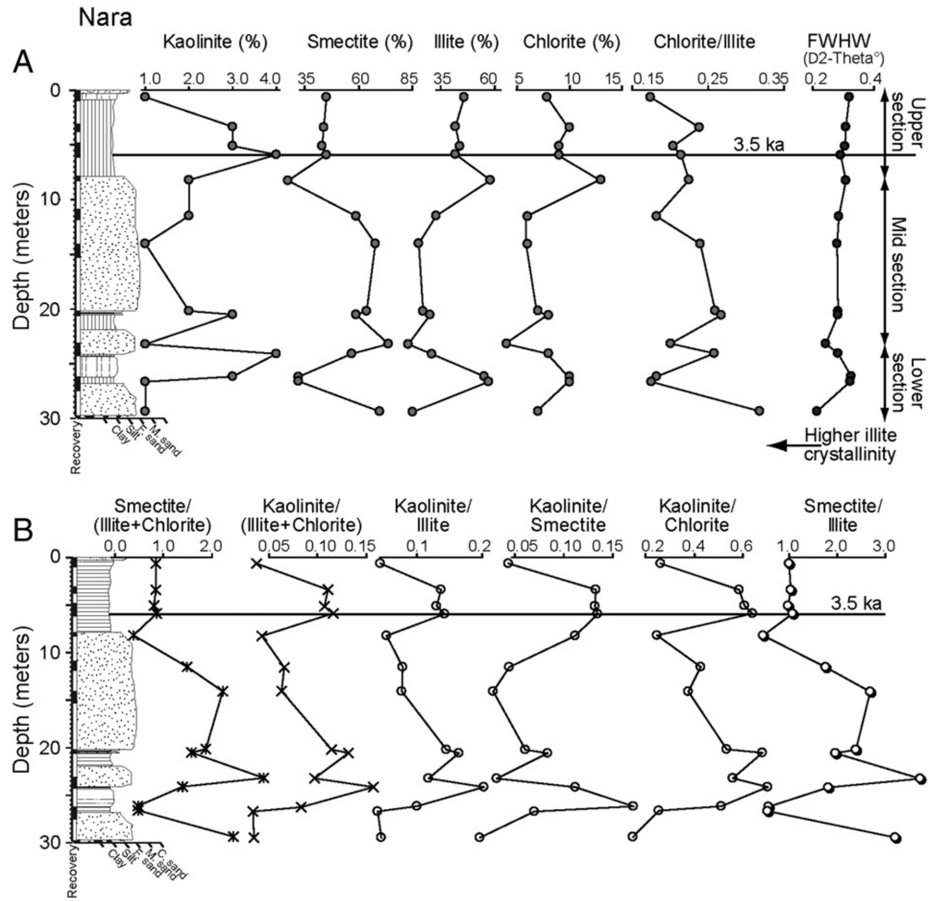

Figure 6. Down-core variation in clay minerals at the Nara borehole showing (A) variations in the relative abundance of kaolinite, smectite, illite and chlorite percentages along with chlorite/illite ratio and the crystallinity index for illite as measured by the FWHW. (B) Variations in climate-sensitive proxy ratios: smectite/(illite + chlorite), kaolinite/(illite + chlorite), kaolinite/illite, kaolinite/smectite, kaolinite/chlorite and smectite/illite.

Links between leaching processes and hydrolization can be assessed by plotting smectite/kaolinite against illite crystallinity (Fig. 8B). Again there is no clear overall relationship but the plot indicates that sediments at different boreholes have different weathering histories. Sediments from Tilwalla, and to a lesser extent the Nara Valley, show the highest smectite/kaolinite values, suggestive of reduced leaching in the presence of the warm, dry environment. This is consistent with their position on the edge of the Thar Desert. Sediments from Marot have moderate smectite/kaolinite values and lower illite crystallinity indicating less hydrolization, but more leaching. Sediments from Keti Bandar, as well as Forts Derawar and Abbas have the greatest degrees of leaching but the least hydrolization. This figure suggests different weathering conditions across the flood plains and contrasting provenances. The character of the Keti Bandar sediments certainly suggests that the rivers of the Tilwalla, Marot and Nara regions are not the dominant sediment contributors to the delta. The similarity between Keti Bandar and Forts Derawar and Abbas is consistent with the conclusion from zircon dating and Pb isotope work that indicates that the dunes of the Thar Desert are largely reworked from mixed Indus sediments in the lower reaches, transported to the NE by eolian processes (Alizai et al., 2011a; Alizai et al., 2011b). These dunes sands are then reworked into the channel sands of at Forts Derawar and Abbas. The sources of the Indus River appear to be dominated by regions experiencing weathering in a wet, tropical setting.

Upper Eastern Punjab

The clay records at Marot and Tilwalla show close similarities in the depth variations of the clay ratios (Figs. 4 and 5). Unfortunately poor age control makes it impossible to test whether the variations are synchronous. There is only one ^{14}C age in the core at Tilwalla, at 45,100 ^{14}C yr BP near the base of the section, although a OSL age at 3.84 m depth suggests sedimentation as young as 4900 yr (Clift et al., in press), with an end to sedimentation shortly after this time. This is close to the age of 7300 ^{14}C yr BP seen near to the top of the section at Marot. The age of the thicker sand bodies in each borehole is not well-known, but is presumed to be early Holocene (7–10 ka) to Pleistocene.

The kaolinite/smectite ratio shows a coherent pattern increasing up-section at both Marot and Tilwalla, with the highest values attained in the uppermost, finest grained layers. Similar trends are also seen in kaolinite/illite and kaolinite/(illite + chlorite), all driven by an up-section increase in kaolinite contents. If we interpret the kaolinite/smectite ratio as a weathering proxy that reflects variations between humid-warm (kaolinite-rich) and more dry and seasonal (smectite-rich) climate conditions (Adatte et al., 2002) we may interpret this trend to indicate warmer wetter conditions during the early Holocene enhancing chemical weathering towards the top of the section (Figs. 4B and 5B). At both sites the underlying sand bodies are characterized by higher values of smectite/illite and smectite/(illite + chlorite) and lower values of kaolinite/smectite, kaolinite/illite and kaolinite/(illite + chlorite).

Variations in the clay ratios appear to closely correlate with changes in lithology, showing that these changes have no clear significance as environmental indicators. For example, smectite/illite shows higher values in sands compared to surrounding clay-rich layers. The same is true of smectite/(illite + chlorite). However, we note that at both sites the smectite/(illite + chlorite) ratios are

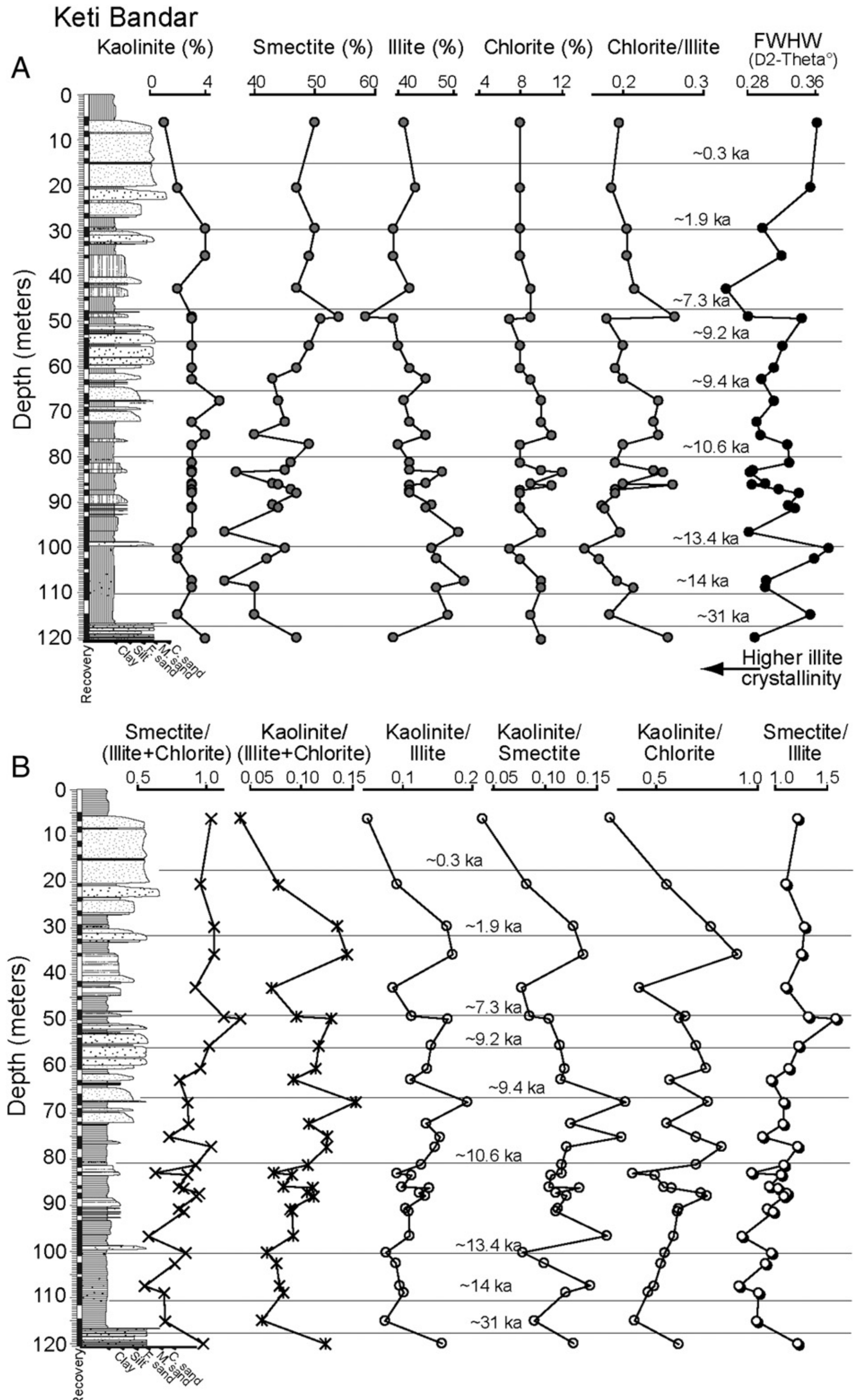

Figure 7. Down-core variation in clay minerals at Keti Bandar showing (A) variations in the relative abundance of kaolinite, smectite, illite and chlorite percentages, along with chlorite/illite ratio and the crystallinity index for illite. (B) Variations in climate-sensitive proxy ratios: smectite/(illite + chlorite), kaolinite/(illite + chlorite), kaolinite/illite, kaolinite/smectite, kaolinite/chlorite and smectite/illite.

lower in the older muds than they are in the younger, shallowest muds, which we interpret to indicate more chemical weathering in recent times compared to the Pleistocene.

Key to this discussion is the origin of the abundant smectite in the Indus flood-plain sediments. The association of high smectite with sands is suggestive of a detrital origin. However, Stern et al. (1997) showed that the oxygen isotopic composition of smectite in older Himalayan Molasse was consistent with a pedogenic origin and that this was even responsive to climate change. Given the predominantly metamorphic and igneous provenance of the Indus sediments smectite-rich detritus from the primary source areas would not be expected, so that a pedogenic origin for smectite in the alluvial

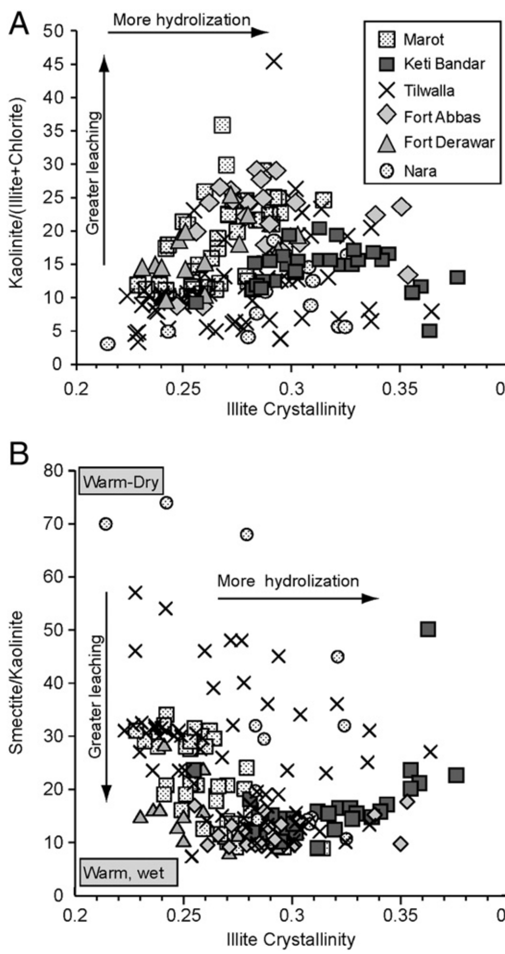

Figure 8. Plots showing the relationship between illite crystallinity and other measures of chemical weathering intensity. (A) Kaolinite/(illite + chlorite) versus FWHM and (B) smectite/(illite + chlorite), neither of which show close correlation with crystallinity.

system seems most probable. Indeed, the generally calcareous nature and slightly alkaline pH values recorded for contemporaneous Mollisols and Entisols on the modern Indus flood plain indicate that present day conditions should favour the formation of smectite. In fact conditions throughout the Holocene are likely to have been conducive to pedogenic smectite formation and therefore the relative increase of smectite in the sands must reflect either a direct or indirect signal related to weathering.

In addition to trends in kaolinite and smectite, illite crystallinity values document a similar trend at both Marot and Tilwalla, with values increasing towards the surface, suggestive of more weathered illite. Both the 'crystallinity' and shape of clay minerals has been used to interpret their origin (Chamley, 1989). This technique works particularly well with illite (Agrawal et al., 1980) and has consequently been widely applied (Chamley, 1989; Pandarinath, 2009). The Indus brings sediments mainly from the glaciated Himalaya and Karakoram and reworked semi-arid to arid soils formed from sediment itself of Himalayan provenance, so that physical/mechanical weathering products (illite and chlorite) dominate the chemical weathering products (smectite and kaolinite) in the river discharge (Rao and Sharma, 1995).

A zone of stronger weathering indicated by increased smectite, increased kaolinite and more weathered illite extends 10–15 m into the subsurface. Our age control allows us to date this transition to the mid-late Holocene, which was a time of weakening summer monsoon rains compared to the early Holocene (Enzel et al., 1999; Fleitmann et al., 2003; Wünnemann et al., 2010). However, this evidence for stronger weathering may reflect the slowing and then end of fluvial transport in the region, rather than less intense summer rainfall, which might otherwise be expected to cause less chemical weathering. Long exposure of clays in the flood plains gives more time for chemical weathering reactions to proceed and this becomes more common in the mid-late Holocene. Even though rates of weathering may reduce the total degree of weathering can increase if transport is sufficiently slow. Climate change expression is frequently recorded by indirect effects and can depend heavily on the topography and climatic zonation of the source regions (Hillier, 1995).

Nara region

Clay mineralogical variability in the Nara Valley sediments is significant and coherent, but is closely linked to lithological variations. Smectite/illite and smectite/(illite + chlorite) are both lower at the top of the section compared to the sand-dominated central part, although we note that the clay-rich parts at 20–27 m depth show even lower values of smectite/(illite + chlorite) (Fig. 6). This is suggestive of less chemical weathering deep in the hole, i.e. before 3.5 ka when we compare similar lithologies. The high smectite/(illite + chlorite) values in the sandier levels would appear to indicate higher degrees of chemical weathering during their sedimentation, but equally this may reflect the source of the sediment as much as temporal variation in the environment. The higher smectite/(illite + chlorite) values in sands compared to clay are consistent with the data from Marot and Tilwalla. In addition, at all sites where smectite values are low, the smectite appears to show a component of mixed layering with illite and is therefore more precisely described as mixed-layer illite–smectite.

Our data show decreasing values of illite crystallinity down-core, indicating more chemical weathering in the shallower clay-rich intervals compared to the deeper sand-rich part of the section. Alternatively, the decrease in smectite and increase in illite, along with the concomitant decrease in illite crystallinity, may reflect increased 'illitization' of the smectite caused by potassium fixation, such as is promoted by cycles of wetting and drying (Srodon and Eberl, 1984).

Because most monsoon reconstructions favour a weakening monsoon since the mid Holocene (<5–7 ka) we suggest that the trends in clay mineralogy observed at Nara and in the Punjabi region may be caused in part by increased cycles of wetting and drying as the summer rains weakened. Smectite/(illite + chlorite) values are generally higher in the Punjab compared to the Nara Valley within the shallowest sediments, indicating more chemical weathering on the flood plains in the north compared to the river further south. Kaolinite/smectite is much lower in the youngest Nara sediment compared to those in the Punjab indicating more leaching in the north and more concentration in a warm and dry climate in the south.

Delta region

The much better age control available at the Keti Bandar site show rapid deposition rates during late Holocene time (Online data supplement: Fig. S2). Two coarsening-upward cycles are represented, separated by a transgressive mud deposited after ~8 ka (Clift et al., 2008), likely correlating with the 8.2 ka flooding event seen in other deltas (Törnqvist et al., 2004). Facies analysis and sequence stratigraphy methods indicate that the delta prograded in two phases at 8–14 ka and 8 ka to present (Giosan et al., 2006).The ages younger than ~13 ka represent deltaic sediments, while older sediments represent the reworking of glacial materials prior to transgression (Clift et al., 2008). Figure 7 shows smectite/(illite + chlorite) ratios increased gradually from ~13 to 8 ka before falling sharply by 6 ka and then continuing at quasi–constant levels to the present day. Kaolinite/

https://doi.org/10.1016/j.yqres.2012.01.008 Published online by Cambridge University Press

smectite ratios show a slightly different trend. This ratio was low at ~13 and ~11 ka, but was higher at ~12 ka and in the early Holocene at 9–10 ka. Kaolinite/smectite shows a decreasing trend after ~7.5 ka (Fig. 7).

We compare these trends with mineralogical results from Kochi (Thamban et al., 2002), a coastal site in western India (Fig. 1), which records run-off from peninsular India, and which is also dominated by a SW monsoonal climate (Fig. 9). Both the Indus delta and Kochi show maximum smectite/(illite + chlorite) around 6–7 ka. However, the Kochi record indicates more chemical weathering only starting after ~9.7 ka, while the increasing trend at Keti Bandar commences at the base of the cored section (~13 ka). Although both broadly correlate with the intensity of the summer monsoon the response is different in each place. In this respect the Indus Delta record parallels trends seen in SE Asia in the Mekong offshore region, which also records smectite/(illite + chlorite) increasing after 14 ka (Colin et al., 2010). This is a period when the monsoon is generally considered to be strengthening (Overpeck et al., 1996; Gupta et al., 2003). Keti Bandar shows a fall in smectite/(illite + chlorite) between 7.3 and 5.9 ka, while Kochi has a single very high value ~6.5 ka. The temporal resolution is low, and it is not clear whether the two sites are really out of step with one another in the timing of the maximum. Smectite/(illite + chlorite) ratios at both sites reach low values at 5–6 ka, indicating weaker chemical weathering, consistent with a reduced summer monsoon at that time. Since ~6 ka to recent times, the smectite/(illite + chlorite) values at both Keti Bandar and Kochi have been variable but lower than the peak in the early Holocene (Fig. 9).

In order to try to understand the linkage between monsoon intensity, chemical weathering and clay mineral formation we plot the clay ratios against select monsoon proxies. Here we compare with a pollen ratio record from Tso (Lake) Kar in Ladakh in northernmost India (Wünnemann et al., 2010), a *Globigerina bulloides* record from the Oman margin of the Arabian Sea (Overpeck et al., 1996) and from a speleothem record from southern Oman, the oxygen isotope character of which is considered to be a robust measure of rainfall intensity (Fleitmann et al., 2003). *Globigerina bulloides* abundance has long been used a proxy for the summer monsoon because the modern abundance of this planktonic foraminifer is closely linked to upwelling induced by summer monsoon winds in that part of the northern Indian Ocean (Prell and Curry, 1981).

It is clear that the monsoon proxies are not all in agreement. The lake records indicate a strengthening of the summer monsoon after 11.4 ka, at the onset of the Holocene, while the upwelling record suggests that the increase began much earlier, ~17 ka. The speleothem record shows summer monsoon strengthening a little later than the lake records after ~10 ka. All the records suggest that the monsoon remained strong and only experienced significant decline after ~6 ka. The clay mineralogy shows a first-order correlation with the monsoon proxies in that it indicates increasing chemical weathering in the Holocene, as the monsoon intensified. However, the Kochi-Cochin record shows a much clearer response to the strengthening after 11 ka than is apparent at Keti Bandar. Furthermore, at both Keti Bandar and Kochi-Cochin the peak in clay mineralogy occurs somewhat after the climatic peak, at 6–8 ka. This suggests a lag of 3000–5000 yr between the change in the climate and a change in the clay mineralogy in the river deltas of the region. Such a delay in the formation of smectite-rich assemblages may reflect the time needed to form clay minerals in the flood plains after the change of climate.

The rapid decline in smectite/(illite + chlorite) ratios in both the deltas after 6–7 ka is not paralleled by any clear sharp decline in the monsoon intensity at that time. The proxies instead point to a gradual weakening of the monsoon then. Indeed the Tso Kar pollen record shows a sharp decline only after 5 ka. Some of this temporal mismatch may reflect uncertainties in the age control at each study

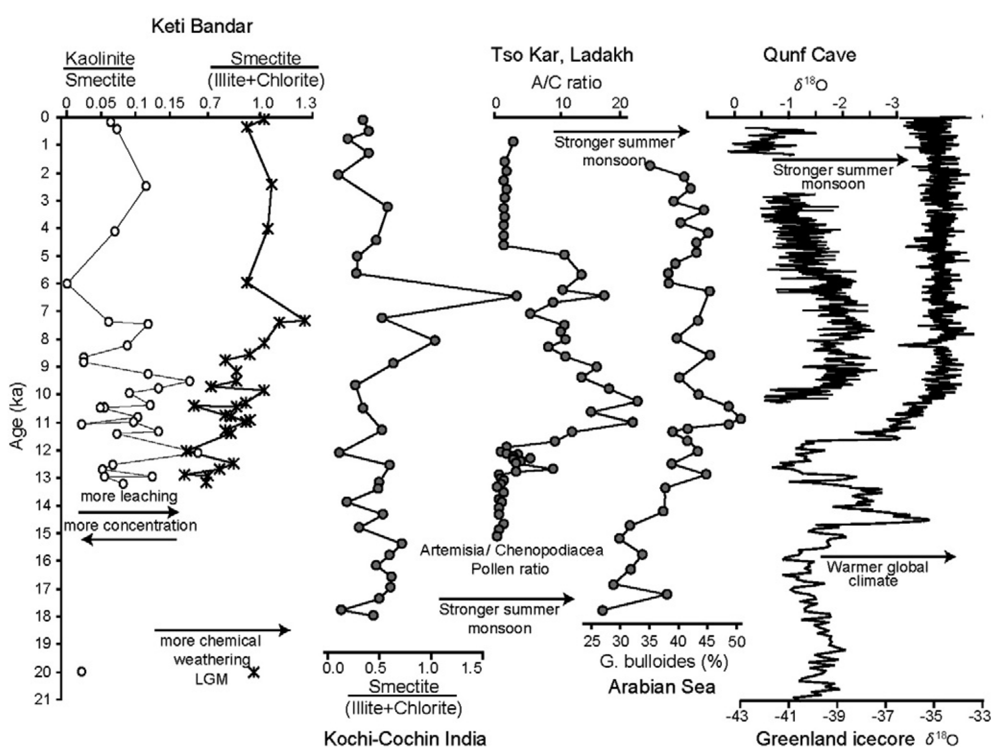

Figure 9. Chart showing the temporal evolution of the environmentally sensitive proxies kaolinite/smectite and smectite/(illite + chlorite) at Keti Bandar compared with that at Cochin-Kochi, India (Thamban et al., 2002). The kaolinite/smectite ratio is used to graph variation in two clays, which are potentially both pedogenic, but indicate different conditions, one (kaolinite) requiring more leaching, the other (smectite) more concentration. We further compare these with three monsoon reconstructions for the Holocene, a moisture-controlled pollen record from Tso Kar in Ladakh (Wünnemann et al., 2010), an upwelling record of Globigerina Bulloides (%) on the Oman margin of the Arabian Sea (Overpeck et al., 1996) and a speleothem record from the Qunf Cave in southern Oman (Fleitmann et al., 2003). The Greenland ice core of Stuiver and Grootes (2000) is shown for comparison.

https://doi.org/10.1016/j.yqres.2012.01.008 Published online by Cambridge University Press

site. We suggest that the delta clay mineralogy does indicate a change in chemical weathering conditions in the river flood plains as the monsoon changes, but that it seems unlikely that clay mineralogy can be used to pin-point environmental changes at high temporal resolution (i.e., <1000 yr). The contrasting responses of Kochi and Keti Bandar also indicate that the marine weathering record will be influenced by the nature of the onshore drainage, especially the degree of sediment buffering and recycling. Erosion of pre-existing fluvial terrace sediments is known to be a significant influence on the flux to the Indus, at least since 10 ka (Bookhagen et al., 2006; Clift et al., 2009).

The clay mineral trends preserved in the delta sediments appear to be consistent with the anticipated weathering response to climate change whereas the floodplain records show an opposing trend. This difference reflects the fact that most of the sediment arriving at the delta is derived directly from Himalayan source regions rather than being reworked on the flood plain. The delta record tends to integrate the overall signal from the catchment and smooth out any spatially variable signatures from the flood plains. Illite crystallinity shows no obvious temporal trend, but it is notable that the values in the delta (max 0.37, min 0.25, mean 0.31) are generally higher than those observed in the floodplain (max 0.36, min 0.21, mean 0.27). This may indicate that the bulk of the sediment delivered to the delta is more weathered than the material we measured from the flood plains in Cholistan (area located to the north of the Thar Desert). This is not consistent, however with the idea that most of the sediment reaching the delta is not derived from this region but represent faster direct transport from the mountains.

Sources and transport of clays in the Indus basin

The clay data from the Indus basin allow us to ask whether clay mineralogy in the delta changes with monsoon intensity. We have discussed the possibility that climate directly influences weathering regimes and thereby the composition of clays being formed or transformed in the flood plains and subsequently transported to the ocean. Another possibility is that climate change causes a change in how and where sediment is eroded from within the basin and that the change in clay assemblage reflects instead, more or less, reworking of sediment previously stored in the flood plains. This latter hypothesis is consistent with the higher smectite/(illite + chlorite) ratios observed in the sands versus the mudstones. Because sands tend to represent either overbank flooding events or channel deposits they are higher energy deposits usually formed by erosion of the flood-plain sediment further upstream, rather than direct delivery from the mountains. This is especially true in the Punjab where the sites are fed by the now ephemeral Ghaggar-Hakra River.

A series of maps for the early, middle and late Holocene (1–6 ka) (Figs. 10A, B and C) show how clay mineral assemblages vary temporally and synchronously across different parts of the flood plain. One feature that is apparent is that the assemblages are quite similar at the broadest scale, with the exception that Nara and Tilwalla are especially rich in smectite. Nonetheless, there are differences across the area that may tell us something of the sediment transport history and where the Indus River derives the bulk of its sediment load. For example, the early Holocene shows that the delta is generally richer in illite compared to the sites in the Punjab or Nara Valley (Fig. 10A). This probably indicates that these regions are not representative of the average clay assemblage in the entire system, which appears to be less chemically weathered than the eastern flood plains.

We suggest that the majority of the clay load of the Indus is dominated by more direct erosion from the Lesser and Greater Himalaya (Clift et al., 2008), as suggested by Nd isotope data rather than reworking from the flood plain. In contrast, in the Ghaggar system more clays were reworked by erosion of the flood plains, where they had been stored and were available for chemical weathering over the period of the LGM. By the mid Holocene (Fig. 10B) the smectite percentage at the delta had increased, but at Tilwalla this clay was less abundant; the sands at Tilwalla have similar high smectite content as those being synchronously deposited at the delta. This is consistent with the idea that reworking of flood-plain sediments is a significant if minority contributor of sediment flux to the ocean during the Holocene. The clays deposited at Nara during the mid and late Holocene are distinctive in their composition compared to the rest of the river system in being more smectite-dominated than seen elsewhere (Fig. 10C). This implies either more chemical weathering in this region or that this region was deriving its sediment from different regions compared to the main Indus, likely with strong influence from the Thar Desert. It is likely that as major channels in the Nara Valley were abandoned chemical weathering increased as transport times slowed, despite the fact that the environment had become drier.

Conclusions

Our present work shows that the clay record reconstructed at the delta (and representing the integrated flux from the Indus River) shows important differences with the terrestrial climate record preserved in the flood plains further upstream, although both reflect changes in weathering and erosion as the monsoon first strengthened, then weakened during the Holocene. The deltaic clay record at Keti Bandar is shown to be a useful proxy record for changing continental weathering environments across the Indus basin despite the fact that the clay mineral proxy record in at least part of the flood-plain shows site-specific trends that are contradictory with the delta record. The Indus delta shows a response to changes in monsoon strength similar to that seen in SE Asia, with more influx of smectite produced by weathering and to a lesser extent more kaolinite during the early Holocene. The Indus record starts its trend to more chemical weathering before that seen in Peninsular India, after ~14 ka compared to 11 ka.

The flood-plain record is harder to interpret because clay assemblages vary strongly with the sediment type, while Keti Bandar is generally muddy. Sands contain more chemically weathered smectite and kaolinite compared to clay-rich layers. This may be because of recycling and reworking, or as a result of differences in the processes affecting the sediment on the flood plain with more wetting and drying, promoting illitization of smectite in environments receiving finer-grained sediment. Furthermore, there is also a possibility of the post-depositional formation of smectite in the sands by peri-pedogenetic or early diagenetic process. Comparison of only clay-rich layers from within a single section shows more smectite and kaolinite going up-section from the mid to late Holocene. This trend, as well as increasing kaolinite/smectite and increasing illite crystallinity, is consistent with more chemical weathering in the flood plain as the monsoon weakened into the late Holocene. This is the reverse of the trend at the delta at that time and shows that this part of the flood plain was not the primary source of sediment to the ocean and/or that the delta record is sufficiently integrative of processes acting across the entire catchment that it is not perturbed by modest local variation. In the Punjabi plains increased weathering probably reflected slower transport as the monsoon weakened, soil-moisture regime became more variable, and sediment flux from the mountains was reduced.

Determining whether the clay mineral assemblages respond immediately to climate change or not is difficult because of poor age control and because of uncertainties about how and when the monsoon changed, depending on which proxy is used. Although there is a general correspondence between clay mineralogy in the largely marine sediments cored at Keti Bandar and monsoon strength the peak response in clays at the delta postdates the climatic peak by 3000–5000 yr. While the monsoon appears to have weakened

https://doi.org/10.1016/j.yqres.2012.01.008 Published online by Cambridge University Press

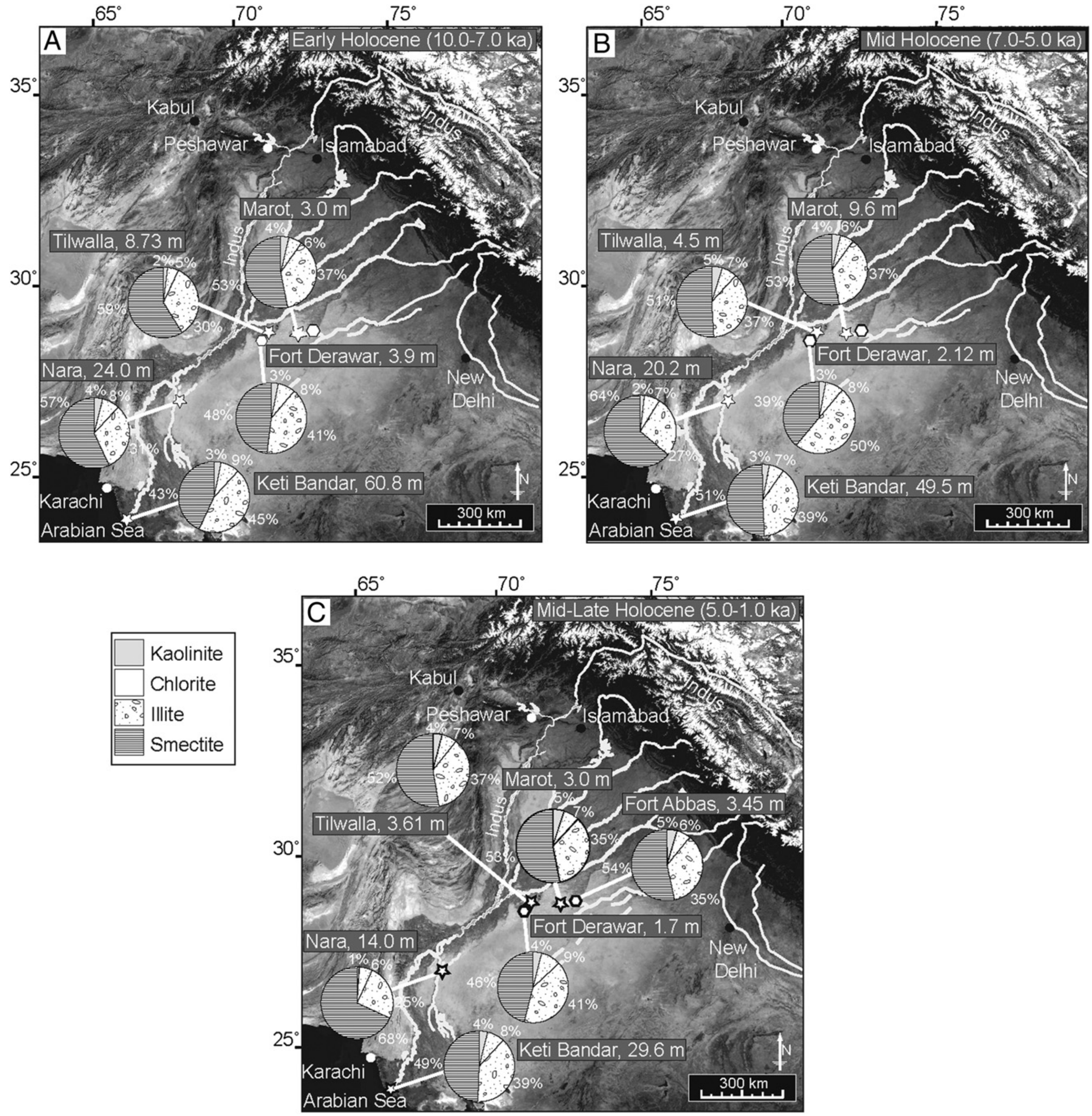

Figure 10. Pie charts showing the estimated composition of clay mineral assemblages across the Indus drainage (A) during the Early Holocene (7–10 ka) (B) during the Mid Holocene (5–7 ka) and (C) during the Late Holocene (1–6 ka).

gradually since the mid Holocene the marine records show a more dramatic fall in smectite and kaolinite after 5–7 ka. Whether this means that clay minerals cannot be used for climate reconstructions on such short time scales or that the present trusted monsoon proxies are not as reliable as we might have hoped is unclear, although we can conclude that clays are not good weathering proxies at time scales of ~1000 yr or less.

Acknowledgments

We are indebted to the Hutton Institute, Aberdeen, for providing facilities from preparation and running of analyses for clay samples. The paper was improved by two careful, anonymous reviewers and we also thank here our editor Alan Gillespie for his helpful comments in improving the quality of the paper. The study was partially supported by a grant from the Leverhulme Trust. PC thanks the Hanse Wissenschaftskolleg, Germany, for the chance to think about these issues. LG acknowledges NSF funding (award OCE-0623766).

Appendix A. Supplementary data

Supplementary data to this article can be found online at doi:10.1016/j.yqres.2012.01.008.

References

Adatte, T., Keller, G., Stinnesbeck, W., 2002. Late Cretaceous to early Paleocene climate and sea-level fluctuations: the Tunisian record. Palaeogeography, Palaeoclimatology, Palaeoecology 178, 165–196.

Agrawal, D., Datta, P., Hussain, Z., Krishnamurthy, R., Misra, V., Rajaguru, S., Thomas, P., 1980. Palaeoclimate, stratigraphy and prehistory in north and west Rajasthan. Journal of Earth System Science 89 (1), 51–66.
Alizai, A., Carter, A., Clift, P.D., VanLaningham, S., Williams, J.C., Kumar, R., 2011a. Sediment provenance, reworking and transport processes in the Indus River by U–Pb dating of detrital zircon grains. Global and Planetary Change 76, 33–55. doi:10.1016/j.gloplacha.2010.11.008.
Alizai, A., Clift, P.D., Giosan, L., et al., 2011b. Pb isotopic variability in the Modern and Holocene Indus River System measured by ion microprobe in detrital K-feldspar grains. Geochimica et Cosmochimica Acta 75, 4771–4795. doi:10.1016/j.gca.2011.05.039.
Birkeland, P.W., 1984. Soils and Geomorphology. Oxford University Press, Oxford. 310 pp.
Biscaye, P.E., 1965. Mineralogy and sedimentation of recent deep-sea clay in the Atlantic Ocean and adjacent seas and oceans. Geological Society of America Bulletin 76, 803–832.
Bockheim, J.G., 1982. Properties of a chronosequence of ultra-xerous soils in the Trans-Antarctic mountains. Geoderma 28, 239–255.
Bookhagen, B., Burbank, D.W., 2006. Topography, relief, and TRMM-derived rainfall variations along the Himalaya. Geophysical Research Letters 33 (L08405). doi:10.1029/2006GL026037.
Bookhagen, B., Fleitmann, D., Nishiizumi, K., Strecker, M.R., Thiede, R.C., 2006. Holocene monsoonal dynamics and fluvial terrace formation in the northwest Himalaya, India. Geology (Boulder) 34, 601–604.
Boulay, S., Colin, C., Trentesaux, A., Clain, S., Liu, Z., Lauer-Leredde, C., 2007. Sedimentary responses to the Pleistocene climatic variations recorded in the South China Sea. Quaternary Research 68, 162–172.
Campbell, I.B., Claridge, G., 1982. The influence of moisture on the development of soils of the cold deserts of Antarctica. Geoderma 28, 221–228.
Chamley, H., 1989. Clay Sedimentology. Springer-Verlag, Berlin. 267 pp.
Clift, P., Giosan, L., Blusztajn, J., et al., 2008. Holocene erosion of the Lesser Himalaya triggered by intensified summer monsoon. Geology 36 (1), 79–82. doi:10.1130/G24315A.1.
Clift, P.D., Giosan, L., Henstock, T., Tabrez, A.R., VanLaningham, S., Alizai, A., Limmer, D., Danish, M., 2009. Sediment buffering and transport in the Holocene Indus River system. American Geophysical Union Eos Trans. AGU, Fall Meet. Suppl., Abstract, T35B-1950.
Clift, P.D., Carter, A., Giosan, L., Durcan, J., Tabrez, A.R., Alizai, A., VanLaningham, S., Duller, G.A.T., Macklin, M.G., Fuller, D.Q., Danish, M., 2012. U-Pb zircon dating evidence for a Pleistocene Sarasvati River and Capture of the Yamuna River. Geology. doi:10.1130/G32840.1.
Colin, C., Siani, G., Sicre, M.-A., Liu, Z., 2010. Impact of the East Asian monsoon rainfall changes on the erosion of the Mekong River basin over the past 25,000 yr. Marine Geology 271 (1–2), 84–92. doi:10.1016/j.margeo.2010.01.013.
Enzel, Y., Ely, L.L., Mishra, S., et al., 1999. High-resolution Holocene environmental changes in the Thar Desert, northwestern India. Science 284, 125–128.
Fagel, N., 2007. In: Hillaire-Marcel, C., Vernal, A.d. (Eds.), Marine clay minerals, deep circulation and climate. : Paleoceanography of the Late Cenozoic, Volume 1. Methods, Elsevier, Amsterdam, pp. 139–184.
Fleitmann, D., Burns, S.J., Mudelsee, M., Neff, U., Kramers, J., Mangini, A., Matter, A., 2003. Holocene forcing of the Indian monsoon recorded in a stalagmite from southern Oman. Science 300 (5626), 1737–1739.
Ghose, B., Kar, A., Husain, Z., 1979. The lost courses of the Saraswati River in the Great Indian Desert; new evidence from Landsat imagery. The Geographical Journal 145 (3), 446–451.
Giosan, L., Clift, P.D., Blusztajn, J., Tabrez, A., Constantinescu, S., Filip, F., 2006. On the control of climate- and human-modulated fluvial sediment delivery on river delta development: the Indus. Eos, Transactions, American Geophysical Union 87 (52) OS14A-04.
Griffin, J., Windom, H., Goldberg, E., 1968. The Distribution of Clay Minerals in the World Ocean. Elsevier, Amsterdam.
Gupta, A.K., Anderson, D.M., Overpeck, J.T., 2003. Abrupt changes in the Asian southwest monsoon during the Holocene and their links to the North Atlantic Ocean. Nature 421, 354–356.
Hamann, Y., Ehrmann, W., Schmiedl, G., Kuhnt, T., 2009. Modern and late Quaternary clay mineral distribution in the area of the SE Mediterranean Sea. Quaternary Research 71 (3), 453–464.
Hillier, S., 1995. Erosion, sedimentation, and sedimentary origin of clays. In: Velde, B. (Ed.), Clays and the environment. Springer Verlag, Berlin, pp. 162–219.
Hillier, S., 2003. Quantitative analysis of clay and other minerals in sandstones by X-ray powder diffraction (XRPD). In: Worden, R.H., Morad, S. (Eds.), Clay Mineral Cements in Sandstones. Special Publication, International Association of Sedimentologists.
Jacobs, M., 1970. Clay mineral investigations of Cretaceous and Quaternary deep sea sediments of the North American Basin. Journal of Sedimentary Research 40 (3), 864–868.
Jeong, G.Y., Hillier, S., Kemp, R.A., 2011. Changes in mineralogy of loess–paleosol sections across the Chinese Loess Plateau. Quaternary Research 75 (1), 245–255.
Lamy, F., Hebbeln, D., Wefer, G., 1998. Late Quaternary precessional cycles of terrigenous sediment input off the Norte Chico, Chile (27.5°S) and palaeoclimatic implications. Palaeogeography Palaeoclimatology Palaeoecology 141, 233–251.
Moore, D., Reynolds, R., 1989. X-ray Diffraction and the Identification and Analysis of Clay Minerals. Oxford University Press, Oxford. 332 pp.
Morgan, R., 1973. The influence of scale in climatic geomorphology: a case study of drainage density in West Malaysia. Geografiska Annaler. Series A. Physical Geography 55, 107–115.
Overpeck, J., Anderson, D., Trumbore, S., Prell, W., 1996. The southwest Indian Monsoon over the last 18000 years. Climate Dynamics 12, 213–225.
Pandarinath, K., 2009. Clay minerals in SW Indian continental shelf sediment cores as indicators of provenance and palaeomonsoonal conditions: a statistical approach. International Geology Review 51 (2), 145–165.
Prell, W.L., Curry, W.B., 1981. Faunal and isotopic indices of monsoonal upwelling: western Arabian Sea. Oceanologica Acta 4, 91–98.
Rao, D.R., Sharma, K.K., 1995. Petrological and geochemical constraints on the petrogenesis of the Jaspa granitic pluton, Lahual region, NW Himalaya. Journal of the Geological Society of India 45 (6), 629–642.
Rateev, M., Gorbunova, Z., 1969. The distribution of clay minerals in the oceans. Sedimentology 13 (1–2), 21–43.
Singer, A., 1984. The paleoclimatic interpretation of clay minerals in sediments—a review. Earth-Science Reviews 21, 251–293.
Srodon, J., Eberl, D., 1984. Illite. Reviews in Mineralogy and Geochemistry 13 (1), 495–544.
Stern, L.A., Chamberlain, C.P., Reynolds, R.C., Johnson, G.D., 1997. Oxygen isotope evidence of climate change from pedogenic clay minerals in the Himalayan molasse. Geochimica et Cosmochimica Acta 61 (4), 731–744.
Stuiver, M., Grootes, P.M., 2000. GISP2 oxygen isotope ratios. Quaternary Research (New York) 53 (3), 277–284.
Thamban, M., Rao, V.P., 2005. Clay minerals as palaeomonsoon proxies: evaluation and relevance to the late Quaternary record from SE Arabian Sea. In: Rajan, S., Pandey, P.C. (Eds.), Antarctic Geoscience: Ocean-atmosphere Interaction and Paleoclimatology. National Centre for Antarctic & Ocean Research, Goa, India, pp. 198–215.
Thamban, M., Rao, V.P., Schneider, R.R., 2002. Reconstruction of late Quaternary monsoon oscillations based on clay mineral proxies using sediment cores from the western margin of India. Marine Geology 186, 527–539.
Thiry, M., 2000. Palaeoclimatic interpretation of clay minerals in marine deposits; an outlook from the continental origin. Earth-Science Reviews 49 (1–4), 201–221.
Törnqvist, T.E., Bick, S.J., Gonzalez, J.L., van der Borg, K., de Jong, A.F.M., 2004. Tracking the sea-level signature of the 8.2 ka cooling event: new constraints from the Mississippi Delta. Geophysical Research Letters 31 (L23309). doi:10.1029/2004GL021429.
Valdiya, K.S., 2002. Saraswati: The River that Disappeared. 1st. University Press, India. Limited, Hyderabad, India, 116 pp.
Wan, S., Li, A., Clift, P.D., Stuut, J.-B.W., 2007. Development of the East Asian monsoon: mineralogical and sedimentologic records in the northern South China Sea since 20 Ma. Palaeogeography, Palaeoclimatology, Palaeoecology 254 (3–4), 561–582.
Wilson, M.J., 1999. The origin and formation of clay minerals in soils: past, present and future perspectives. Clay Minerals 34 (1), 7–25.
Wünnemann, B., Demske, D., Tarasov, P., et al., 2010. Hydrological evolution during the last 15 kyr in the Tso Kar lake basin (Ladakh, India), derived from geomorphological, sedimentological and palynological records. Quaternary Science Reviews 29, 1138–1155.
Zimmermann, H.B., 1977. Clay–mineral stratigraphy and distribution in the South Atlantic ocean. Initial Reports of the Deep Sea Drilling Project 39, 395–405.