Weathering in the Himalaya, an East- West comparison: Indications from major elements and clay mineralogy

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Abstract

Studying past weathering regimes is important for a better understanding of the influence of climate on weathering, erosion and runoff. The Himalayan foreland basin contains a record of tectonics and paleo-climate since Miocene times. Spanning the entire mountain range, the Mio-Pliocene detrital Siwalik Group allows studies to directly compare the western and eastern Himalaya within similar sedimentary settings. In this study, we use major elements and clay mineralogy to reconstruct the weathering regime along strike since Miocene times. We studied previously dated Dharamsala (pre-Siwalik) and Siwalik sections in the western (Joginder Nagar, Jawalamukhi and Haripur Kolar sections) and eastern (Kameng section) Himalaya in order to constrain variations in weathering regimes along strike. The compilation of the three sections in the west makes for one of the longest continuous sedimentary records in the Himalaya, spanning over 20 Ma. The K/Al ratio is used as a reliable weathering proxy and shows a trend toward more intense weathering over time in both the west and the east, but with sediments in the western Himalaya generally more weathered than in the east, despite higher precipitation in the east. Clay minerals and major elements indicate similar lateral variations in weathering. More intense weathering in the west is linked to a more seasonal climate, permitting weathering of sediments during the dry season, whereas higher runoff in the east leads to more rapid erosion and sediment transport, inhibiting extensive weathering.

1. Introduction

The Himalayan mountain belt, together with the Tibetan plateau, exerts a strong influence on regional climate, acting as an orographic barrier for the Asian monsoon (Boos and Kuang, 2010). The monsoonal climate, in turn, has a major influence on erosion and relief patterns (Bookhagen and Burbank, 2006; Clift et al., 2008; Thiede et al., 2004), which influence chemical weathering intensity and fluxes (Galy and France-Lanord, 2001; West et al., 2005). Chemical weathering plays a central role in global CO_2 drawdown and climatic cooling since the early Cenozoic (e.g., Kump et al., 2000). Thus, the Himalaya play a central role in the globally coupled tectonic – climate – erosion system, and studies of past weathering rates and regimes are crucial to unravel interactions between tectonics, erosion, climate and weathering (e.g., Derry and France-Lanord, 1996). Moreover, spatial variations in tectonics, erosion and weathering patterns can have implications for the past climate and the evolution of the mountain belt. Lateral variations in erosion and exhumation rates in the Himalaya have been investigated (e.g., Galy and France-Lanord, 2001; Thiede and Ehlers, 2013; van der Beek et al., 2016), but studies on spatial and temporal variations of the weathering regime and intensities remain rare.

The modern monsoonal climate strongly impacts precipitation patterns in the Himalayan region. Monsoonal winds take up moisture in the Arabian Sea and the Bay of Bengal and transport it towards the Himalayan mountain front. This results in strong precipitation during the northern-hemisphere summer months. The precipitation pattern varies along strike, with generally more precipitation in the east than in the west (Bookhagen and Burbank, 2010). During the winter months, precipitation is mostly focused on the western and eastern terminations of the mountain belt, but the amount of precipitation remains higher in the east than in the west (Bookhagen and Burbank, 2010). Himalayan river discharge and sediment transport to the sea is linked to precipitation, and has an impact on the storage of sediment in the floodplain (Andermann et al., 2012a; 2012b). Lupker et al. (2012) showed that sediments in the floodplain are more weathered than sediments collected from Himalayan rivers at the mountain front, pointing out the important role of chemical weathering in the floodplain.

The Himalayan foreland-basin sediments, together with offshore sediments in the Indus and Bengal fans, hold a record of tectonics, erosion, and climate since Miocene times. Numerous studies have aimed at reconstructing paleo-vegetation and monsoon evolution from this record (e.g., Clift et al., 2010; France-Lanord and Derry, 1994; Freeman and Colarusso, 2001; Quade et al., 1989; Vögeli et al., in review). The most recent studies on the onset of the monsoon date it back to the Eocene (Licht et al., 2014), even though the evolution of the monsoon and its impact on precipitation patterns remains to be discussed. A change in vegetation, from C3- to C4-plant dominated, has been documented at ~7 Ma, which was interpreted as indicating drying of the climate as it became more seasonal (Dettman et al., 2001).

Continental Himalayan foreland-basin sediments of the Siwalik group crop out along the entire mountain front, allowing west-east comparisons within similar sedimentary settings, while more scattered pre-Siwalik deposits allow pushing the record back to Early Miocene times. Here we present paleo-weathering data of newly sampled Dharamsala (pre-Siwalik) and Siwalik sections in north western India (Figure 1); the compilation of these sections makes for the longest continuous sedimentary record in the Himalayan foreland basin. We use clay mineralogy and whole-rock geochemistry to reconstruct the weathering intensity and compare the Siwalik sections in the western Himalaya with the Kameng River section in the eastern part of the Himalayan foreland basin Vögeli et al., accepted). Lateral differences in $\delta^{13}C$ of organic matter of bulk sediments from these same sections have been interpreted as recording differences in the evolution of vegetation from west to east (Vögeli et al., in review b). The current study is aimed at better understanding the relationship between changes in vegetation, climate and weathering regime, by directly comparing the evolution of the weathering regime in the west and the east, using the same proxies.

2. Geological setting

The evolution of the Himalaya is mainly driven by the early Cenozoic collision of the Indian and Eurasian continents (X. Hu et al., 2016), which resulted in major crustal shortening and thickening (Hodges, 2000; Yin and Harrison, 2000). A north-dipping fault system separates the Himalaya into four major litho-tectonic units (Figure 1), which are, from north to south: the Tethyan Sedimentary Series (TSS), the Higher Himalayan Crystalline Series (HHCS), the Lesser Himalayan Series (LHS) and the Sub-Himalayas (SH). The main faults are, from north to south: the South Tibetan Detachment System (STDS), which separates the TSS from the HHCS; the Main Central Thrust (MCT), which lies between the HHCS and the LHS; the Main Boundary Thrust (MBT), which separates the LHS and the SH; and the Main Frontal Thrust (MFT), which thrusts the SH over the Ganga/Brahmaputra plain (DeCelles et al., 2001; Le Fort, 1986; Yin and Harrison, 2000). The TSS is a Paleozoic-Eocene sedimentary succession that was deposited on the Indian passive margin (Gaetani and Garzanti, 1991). The HHCS comprises high-grade metamorphic rocks and granites, whereas the LHS is composed of low-grade metasedimentary rocks (Hodges, 2000). The LHS has been subdivided into the Inner Lesser Himalaya (iLH) and the Outer Lesser Himalaya (oLH), based on different Nd-isotopic signatures (Ahmad et al., 2000): the iLH is characterized by strongly negative ε_{Nd} values, whereas the oLH has ε_{Nd} values similar to the HHCS and represents the low-grade metamorphic cover of the latter. The Sub-Himalaya, the outermost unit, consists of the deformed Mio-Pliocene foreland-basin deposits of the Siwalik Group, and crops out along the Himalaya from Pakistan all the way to northeastern India (DeCelles et al., 1998; Ojha et al., 2009) (Figure 1).

Pre-Siwalik continental Cenozoic foreland-basin sedimentary rocks are found in the western Himalaya; they are termed the Dharamsala Group and represent the Late Oligocene/Early Miocene infill of the Himalayan foreland basin (Raiverman et al., 1983). They consist of continental fluvial, lacustrine or deltaic sediments, and contain fine-to medium grained sandstones, siltstones and overbank mudstones containing soil-carbonate nodules (Najman et al., 2004). The Siwalik Group is divided into the Lower, Middle and Upper Siwalik sub-groups, which are also known by local and laterally varying formation names. The Lower Siwaliks (LS) consist of mudstones with some development of paleosols, alternating with fine- to coarse-grained sandstones. The abundance of paleosols varies laterally along strike and decreases towards the east. The LS were deposited by high-sinuosity streams (Nakayama and Ulak, 1999). The Middle Siwaliks (MS) are characterized by massively bedded, medium- to coarse-grained micaceous sandstones. The MS represent a depositional environment of large braided rivers. The Upper Siwaliks (US) consist of beds of conglomerates alternating with sandstone beds, deposited by gravelly braided rivers. Mudstones and paleosols are less frequent. Overall coarsening upward is observed in the entire Siwalik Group, resulting from forward propagation of the Himalayan thrust front

(DeCelles et al., 1998; Dubille and Lavé, 2015), and boundaries between the sub-groups are gradual.

The three sampled sections of this study in Himachal Pradesh, northwestern India, contain Dharamsala (Joginder Nagar) and Siwalik (Joginder Nagar, Jawalamukhi and Haripur Kolar) sedimentary rocks. They were previously dated by magnetostratigraphy (Meigs et al., 1995; Sangode et al., 1996; White et al., 2001) and provide a composite age record that ranges from 20 to 1 Ma (Figure 2). The Joginder Nagar (JN) section ranges from ~20 to 12 Ma and contains rocks of the Lower and Upper Dharamsala and Lower Siwalik subgroups, with boundaries for the Lower/Upper Dharamsala at 16.5 Ma and Upper Dharamsala/LS at 12.5 Ma (White et al., 2001). In the Jawalamukhi section (JW), all three Siwalik sub-groups (LS, MS and US) are present, with boundaries set at 10.9 Ma between LS and MS and at 6.8 Ma for the MS/US boundary (Meigs et al., 1995). The Haripur Kolar section (HK) contains MS and US with the MS/US boundary at 5.23 Ma (Sangode et al., 1996). Soil-carbonate nodules are present in Siwalik sediments in Pakistan (Quade et al., 1989), northwestern India (Sanyal et al., 2010), and western and central Nepal (DeCelles et al., 1998; Quade et al., 1995). They have never been reported more eastwards and are lacking in the Kameng River section in Arunachal Pradesh, northeastern India (Vögeli et al., accepted). The setting of the eastern Himalayan Siwalik section (Kameng section; KM) has been described recently by Chirouze et al (2013) and Vögeli et al. (accepted).

3. Sampling and methods

3.1 Sampling strategy

In order to maximize the age constraints, we sampled according to the magnetostratigraphic sampling points of White et al. (2001) for the JN section, Meigs et al. (1995) for the JW section, and Sangode et al. (1996) for the HK section, using field notes and maps of these previous publications. 2-3 samples per Ma were collected to obtain a continuous age record. Samples were collected in pairs of adjacent fine- (mud/siltstone) and coarse-grained (sandstone) sediment beds of the same age; fine-grained samples were sampled in paleosols where present. Additionally, soil-carbonate nodules were collected where present, which were analyzed for stable isotopes (Vögeli et al., in review b). Modern river sand and mud was collected in proximity to the sections.

3.2 Methods

3.2.1 Clay mineralogy

The <2 μm fraction of clay minerals was extracted from selected samples from the JN, JW and HK sections. To remove carbonate and organic matter, the samples were treated with 1M acetic acid and dissolved $Na_4P_2O_7$, respectively. Samples were cleaned with MilliQ water after each removal. The <2 μm fraction was separated by centrifuging the samples (diluted in MilliQ) for 8 minutes at 700 rpm and pumping off the top 7 cm of the suspended fraction. This procedure was repeated until a volume of 2 liters was reached (Moore and Reynolds, 1997). Oriented aggregates were made on glass slides. X-ray diffractograms were carried out on air-dried and ethylene-glycol-treated samples on a Bruker D8 Advance X-Ray Diffractometer at ISTerre, Université Grenoble-Alpes.

Clay-mineral assemblages were obtained by a semi-quantitative peak analysis of the XRD patterns. Based on their peak heights, clay minerals were added up to 100% (Capet et al., 1990), obtaining a percentage of different clay minerals with a relative error of ~5% (Holtzappel, 1985). Illite crystallinity, also known as the Kübler Index (KI), was obtained by measuring the full width at half maximum of the illite 10 Å peak on an X-ray diffractogram. The KI determines three zones of very low-grade metamorphism: diagenesis, anchizone and epizone (Kübler and Jaboyedoff, 2000).

Clay-mineral assemblages can be used as an indicator for weathering regimes (Setti et al., 2014). Smectite forms as a secondary clay mineral in soils and is enhanced in seasonal and warm climates (Hillier, 1995; Huyghe et al., 2005; 2011), although formation of smectite is also favored by weathering of volcanic rocks (Chamley, 1989). Kaolinite is preferentially formed in warm and humid climates (Righi and Meunier, 1995; Setti et al., 2014). Smectite and kaolinite should become more abundant with increasing weathering intensity. Illite and chlorite represent detrital clays resulting from the physical erosion of the Himalayan range, although illite can also be formed diagenetially; the KI (see above) allows discriminating between detrital and diagenetic illite. Sediment provenance can potentially influence clay mineralogy (Chamley, 1989; Garzanti et al., 2014) and should therefore be taken into account when interpreting clay-mineral assemblages in terms of weathering.

3.2.2 Major elements

Samples were ground to a powder in an agate mortar, after which 50-70 mg of powder was dissolved in a mixture of HF and HNO_3 and heated for 72 h at 90 °C. The solution was subsequently treated with boric acid to neutralize acids, and H_2O_2 to dissolve organic matter. Major-element concentrations were analyzed on a Varian 720-ES inductively coupled plasma atomic emission spectrometer (ICP-AES) at ISTerre, Université Grenoble Alpes, using the method of Chauvel et al. (2011). International standard reference material was analyzed parallel to the samples and was used to evaluate the accuracy at ~3 %, by comparing measured and reference values. Loss on ignition (LOI) was obtained by weight loss after heating at 1000 °C for an hour. Hydration (H_2O^+) of selected samples was measured at the Service d'Analyse des Roches et des Minéraux (SARM), Centre de Recherches Pétrographiques et Géochimiques (CRPG) in Nancy, France, with the Karl Fischer titration method (cf. Lupker et al., 2012).

Ratios of mobile to immobile elements (K/Al, K/Si) and sediment hydration (H_2O^+) were used as proxies for weathering intensity. However, ratios of K/Si and H_2O^+/Si are primarily controlled by grain size and only secondarily by weathering (Lupker et al., 2012; 2013). When plotting K/Si against Al/Si, coarse-grained sediments are represented in the lower Al/Si range (< 0.15), and the steepness of the regression line is controlled by the degree of weathering. Normalization of the mobile-to-immobile element ratios allows corrections for differences in grain size of different samples. In order to obtain grain-size independent proxies, ratios were normalized (K/Si* and H_2O^+/Si*) to a common Al/Si of 0.22 (the average Al/Si of all measured coarse-grained samples in the west and east), with a regression line through a coarse-grained end-member and the sample, following Lupker et al. (2013). The K/Al ratio was also used as a more direct proxy for chemical weathering intensity, as K/Al seems not to be primarily controlled by grain size (D. Hu et al., 2016).

4. Results

Here we report clay-mineral assemblages and major-element data for the Joginder Nagar, Jawalamukhi and Haripur Kolar sections in northwest India. These are subsequently compared to similar data from the eastern Himalaya (Kameng river section; Vögeli et al., accepted).

4.1 Clay mineralogy

Clay minerals in the <2 μm fraction of clayey beds in the three western sections consist mainly of illite, chlorite, smectite and kaolinite, the relative proportions of which vary with time. In the JN and the lower part of the JW sections (Dharamsala and LS deposits dated from 20 to ~10.5 Ma), clays are dominated by illite (illite+chlorite/∑clays > 0.6), which is followed by an interval in which smectite and kaolinite are dominant (illite+chlorite/∑clays < 0.5). Samples with depositional ages between ~8 and 5 Ma have particularly high smectite content (> 50%). From 4 to 1 Ma, illite concentrations increase again, but smectite remains more abundant than in the lower part of the section. The modern Beas river sample is dominated by illite (65%; Figures 2, 3).

Illite crystallinity (KI) is fairly constant throughout the three sections (Figure 3). Values vary from 0.12 to 0.23 $\Delta°2\theta$ with illite crystallinity similar to muscovites from the Higher Himalayan Crystalline Series (Huyghe et al., 2005). These values are representative for the epizone of low-grade metamorphism, and indicate that illites are detrital rather than diagenetic.

4.2 Major elements

Pairs of coarse- and fine-grained samples were analyzed; complete major-element results are provided in Supplementary Table S1. Major-element compositions reflect the difference in grain size, with coarse-grained samples containing more SiO_2, whereas finer sediments have higher K_2O and Al_2O_3 concentrations. The ratio of the immobile elements Al_2O_3/SiO_2 can be used as a grain-size proxy (Lupker et al., 2012). Plots of the concentrations of SiO_2 vs K_2O and Al_2O_3 show a separation of fine- and coarse-grained sediments (Figure 4). Ratios of mobile to immobile elements, such as K/Al, are used to quantitatively track weathering intensity over time (Figure 5). K/Al ratios in the western sections are relatively constant at ~0.2 over the last 20 Ma, with an excursion to higher values of ~0.4 between ~7 and 5 Ma. The concentration of calcium is very variable in the western sections, with an average of 3.7 wt % oxide (Figure 6). CaO does not correlate with SiO_2, and therefore does not appear to be dependent on grain size.

5. Discussion

Lateral variation in the climatic evolution of the Himalaya is reflected in a varying evolution of vegetation, as inferred from the record of stable carbon isotopes, with C4 plants becoming dominant in the western Himalaya, but not in the east, around 7 Ma. The vegetation in the eastern part of the orogen is dominated by C3 plants since Miocene times (Vögeli et al., in review b). The dominance of C4 plants since ~7 Ma in the western Himalaya is interpreted as resulting from the change to an overall drier and more seasonal climate at that time (Dettman et al., 2001; Quade and Cerling, 1995; Quade et al., 1989). In contrast, climate in the east has remained too humid to develop C4 vegetation (Vögeli et al., in review). By using complementary proxies to stable carbon isotopes, such as clay minerals and major elements, the influences of this difference in climatic evolution on weathering intensities and regimes are investigated here. Inferences on weathering intensity and regime also depend, however, on the sensibility of the different proxies. In the following, we first discuss potential provenance and diagenetic influences on weathering signals in the western sections, and then compare the west and the east.

5.1 Provenance and diagenesis in the western sections

The provenance of sediments from the Joginder Nagar section has been determined using whole-rock Sm-Nd isotopes, detrital white-mica Rb-Sr and Ar-Ar ages (White et al., 2002), and detrital monazite U-Pb ages (White et al., 2001). Najman et al. (2009) used Ar-Ar ages on detrital white micas, sandstone petrography and Sr-Nd isotopic compositions in the Jawalamukhi section to reconstruct changes in provenance. The Lower Dharamsala (~20-17 Ma) sediments are mainly sourced from the HHCS (Figure 2). At the boundary from the Lower to Upper Dharamsala, provenance changes from high-grade metamorphic rocks to low-grade metamorphic and sedimentary rocks of the oLH (Haimanta series; White et al., 2002). iLH material is present in the Siwalik sediments from at least ~9 Ma. In the Jawalamukhi section, HHCS material cuts out completely at 7 Ma (Najman et al., 2009) and the iLH becomes the dominating source of the Siwalik sediments. From 6 Ma on, detritus from Proterozoic granitoids is present. Isotopic provenance studies in the Haripur Kolar section are lacking. Basic provenance analysis was based on clay minerals and petrography and indicates an HHCS and LHS source (Suresh et al., 2004). Peak activity of the Main Frontal Thrust has been established at 1.8 Ma, and is associated with recycling of Siwalik sedimentary rocks in the uppermost Siwalik series (Kumar et al., 2003). Major changes in provenance thus occurred at ~17 Ma (oLH coming in), at 9 Ma (iLH coming in), and at 7 Ma (HHCS cuts out).

Illite crystallinity (Kübler Index) can be used to distinguish different low-grade metamorphic zones. In the Karnali section in western Nepal, illite crystallinity starts to increase from a stratigraphic depth of ~2100 m (Huyghe et al., 2005), indicating diagenetic illite at these depths. Illitisation of smectite begins at a temperature of 70-95°C (Dunoyer De Segonzac, 1970). It is thus important to approximately know to what temperatures the rocks in our sections could have been heated during burial, in order to establish the potential diagenetic influence on other proxies. Additionally to illite crystallinity, apatite fission-track (AFT) ages can be used to constrain a burial temperature. Partially or fully reset AFT ages are a clear indicator that sedimentary rocks were heated to 60-120°C, the AFT partial annealing zone (Tagami and O'Sullivan, 2005). Estimated burial depths for total AFT annealing are ~4000 m in the Kameng section (Chirouze et al., 2013) and ~3000 m in central/western Nepal (Surai, Tinau, Karnali sections; van der Beek et al., 2006), respectively. AFT analyses are lacking for the JN, JW and HK sections, but paleotemperature estimates from vitrinite reflectance of the Dagshai (max. age 33 Ma) and Kasauli (max. age 22 Ma) formations, which are the along-strike equivalents of the Dharamsala Group further east, are between ~160 and 200 °C (Najman et al., 2004). In contrast, in the western sections (this study), illite crystallinity is relatively constant and lies in the epizone field, suggesting that the illite is mostly detrital, similar to what is observed in the Kameng section (Figure 3). Illite crystallinity therefore reflects source-area metamorphism. None of the western samples fall into the diagenesis field, which should be the case if illitisation of smectite had occurred (Huyghe et al., 2005; Lanson et al., 1995). There is no sign of illitisation of smectite throughout the western sections, suggesting that the influence of diagenesis is likely to be minor, or at least not affecting the clay minerals in the western sections.

In these sections, clay mineralogy begins to change at ~11 Ma, when smectite becomes more abundant and remains so until ~1 Ma. Clay mineralogy, therefore, does not appear correlated with changes in provenance, as smectite becomes abundant at ~11 Ma and remains stable after that, whereas provenance changes occur at ~9 Ma and especially at 7 Ma. Since neither diagenesis nor provenance are likely to bias the clay-mineralogy signal, variations in clay mineralogy can be mostly ascribed to changes in weathering intensity and regime through time.

K/Al ratios do not show a correlation with burial depth; diagenesis is therefore likely to be of negligible influence on K/Al ratios. K/Al ratios in the west remain relatively constant over time, except between ~7 and 5 Ma, where K/Al values are anomalously high. Several samples between 7 and 5 Ma have K/Al ratios of ~0.4 (Figure 5), indicating the presence of "fresh", nearly unweathered material. These samples were collected in the upper part of the Jawalamukhi section (MS), where conglomerate is frequent (Figure 2). They are depleted in Al because of the grain-size effect. Moreover, conglomerates represent a proximal depositional environment close to the mountain front, where sediments are less weathered compared to those in the floodplain (Lupker et al., 2012). The US of the Haripur Kolar section do not show high K/Al values, high values are only present in sedimentary rocks of the Jawalamukhi section. Provenance changes between 7 and 6 Ma, when material sourced from Proterozoic granitoids are brought in and HHCS is cut out, could potentially influence K/Al ratios because these granitoid sources are expected to have elevated K-concentration. However, the K-concentration in these samples does not show exceptionally high values (Table S1). The increase in K/Al between ~7 and 5 Ma is therefore more likely to be related to the depositional environment becoming locally more proximal and an additional change in provenance, than to a change of climate.

5.2 West-east comparison of weathering regimes

The three pre-Siwalik and Siwalik sections in the western Himalaya form the longest composite continuous sedimentary record in the Himalayan foreland basin, which allows us to reconstruct weathering history since the Early Miocene. Weathering intensity in the western Himalaya is fairly constant from ~21 to ~13 Ma, after which it slightly decreases until 7 Ma, as inferred from K/Al and K/Si* ratios (Figure 5). The period between 7 and 5 Ma is characterized by exceptionally low weathering, reflected in a high K/Al ratio. From 5 to 1 Ma, weathering is relatively constant at a K/Al value of ~0.20, similar to the period from 21 to 13 Ma, with some variation around 3 Ma (Figure 5).

In the eastern Kameng section, K/Al ratios indicate an increase in weathering intensity with time, the Upper Siwaliks being more weathered than the Lower Siwaliks. As the Kameng River section only spans the last 13 Ma, a direct comparison can only be made for the mid-Miocene to Pleistocene. Sediments of the Dharamsala Group in the west (older than 13 Ma, and absent in the east), are generally more weathered than the Lower Siwaliks in the east. In general, sediments from the east show a higher K/Al ratio, hence are less weathered, than sediments from the western sections. The period between ~7 and 5 Ma is exceptional, with sediment in the west being less weathered, showing anomalous values of K/Al up to 0.4. However, as discussed above, we suggest this to be a signal related to the source and the specific depositional environment, rather than regional weathering intensity.

A potential factor controlling variations in weathering intensity could be recycling of Siwalik or older sediments. The Siwaliks show striking differences in width and internal deformation from east to west, with western sections generally being wider and showing more internal thrusts (e.g., Hirschmiller et al., 2014). If such internal deformation led to recycling of Siwalik sediments, this might explain the stronger weathering intensity recorded in the western sections. However, petrographic data from the Joginder Nagar and Jawalamukhi sections do not indicate significant recycling (White et al., 2002; Najman et al., 2009), while in the eastern Kameng section some outlier datapoints were interpreted as indicating limited recycling (Vögeli et al., accepted). In general, significant recycling should lead to increasing weathering intensity upsection, which is observed in the Kameng but not in the western sections. Therefore, we do not think that sediment recycling can explain the differences between the western and eastern sections.

By plotting K/Si against Al/Si (Figure 7), a weathering signal can be extracted from the steepness of regression lines (Lupker et al., 2013). We calculated the 95% confidence interval for each linear regression, to show the uncertainty of the regression line. Confidence levels of regression do not show a clear separation, although eastern sediments seem to be slightly more weathered, which contradicts the inference from the K/Al data and may be due to the period of very low weathering in the west between 7 and 5 Ma (Figure 7, orange circle). By taking out the anomalously unweathered samples between 7 and 5 Ma, we would expect the regression line of the western sections to change its steepness, but it remains steeper than that of the eastern sediments (i.e., indicating less intense weathering). Plotting K/Si versus Al/Si shows the primary control of grain size. Coarse-grained sediments of the western sections are more weathered than coarse-grained sediments in the Kameng; this slight difference influences the steepness of the regression line. In order to directly compare weathering in the west and east, plotting K/Al versus age thus appears more suitable (Figure 5), as absolute K/Al values can be directly compared within different sections. K/Al appears to be a robust proxy for reconstructing weathering intensities, as also confirmed by a recent study in the South China Sea (D. Hu et al., 2016).

Other weathering proxies, such as the Chemical Index of Alteration (CIA) (Nesbitt and Young, 1984) are more difficult to use, due to the variation in carbonate content of the sediments and changes in provenance. Calcium concentrations vary laterally, with higher Ca concentration in the west (average of 3.7 wt % oxide compared to 0.7 wt % oxide in the east) resulting in the abundance of soil-carbonate nodules in the western sections (Figure 6). In contrast, soil-carbonate nodules are absent in the Kameng section; they have never been reported more eastwards than eastern Nepal. As the climate is and probably has been more humid in the past in the east than in the west (Bookhagen and Burbank, 2006; Vögeli et al., in review), it is likely that calcium was dissolved and the formation of soil-carbonate nodules was inhibited in the eastern sections.

$\delta^{13}C_{org}$ shows important variations from west to east along the Himalayan front at ~7/8 Ma (Vögeli et al., in review), which are not reflected in the clay mineralogy. Clay mineralogy within the different Siwalik sections is quite similar, with clay-mineral assemblages starting to be more smectite rich somewhere between 11 and 7 Ma (the exact timing varying laterally), and being characterized by a high illite content in the stratigraphically lower series (Dharamsala and LS). This pattern is also observed in the distal Bengal-Fan record (France-Lanord et al., 1993). In the Kameng River section, the smectite-rich period occurs from 8 Ma until 3 Ma, when kaolinite becomes more abundant. On the other hand, smectite starts to be more abundant at ~11 Ma in the west and stays the dominant clay mineral up to 1 Ma. In the east, the smectite-rich period also coincides with a change in source (Chirouze et al., 2013; Vögeli et al., accepted). However, although sources in the west and the east are different, types of smectite seem to be similar, as indicated by similar XRD peaks (Appendix 2). We therefore suggest that the abundance of smectite is likely to be controlled by climate rather than by provenance. Kaolinite is less abundant in the west than in the east. The modern clay-mineral assemblages of the Beas River (sampled in proximity of the section) in the west is dominated by illite, similar to other Himalayan rivers (Kameng and Subansiri rivers) and the Brahmaputra in the east (Vögeli et al., accepted). In contrast, the Ganges (further into the floodplain) carries more smectite (Huyghe et al., 2011). Smectites can be formed under enhanced weathering and seasonal conditions, as is the case in the Ganges floodplain today. The increasing abundance of smectite since 7-11 Ma can, therefore, be interpreted to reflect a change towards a more seasonal climate (Huyghe et al., 2005). The dominance of smectite in the western sections between 8 and 5 Ma could thus be an indicator of a more seasonal climate, which is supported by the appearance of C4 plants during this time (Quade and Cerling, 1995; Vögeli et al., in review); C4 plants are known to be more resistant to water stress, hence seasonal climate (Ehleringer, 1988). Clay-mineral assemblages and stable carbon isotopes, hence vegetation, thus both reflect mostly seasonality of the climate. Clay mineralogy also suggests increasing weathering intensity with the abundance of smectite at ~8 Ma. K/Al and K/Si ratios, in contrast, suggest less weathering in the period between ~7 to 5 Ma in the west (Figures 5 and 7). In the Kameng section, an overall increase in weathering intensity is observed, with a major change towards more intense weathering at 8 Ma (Figure 5; Vögeli et al., accepted).

Generally, K/Al ratios show that sediments deposited in the east (Kameng section) were less weathered than in the west since the mid-Miocene. In contrast, modern precipitation is higher and less seasonal in the eastern Himalaya, driving more intense runoff and erosion (Bookhagen and Burbank, 2010; Galy and France-Lanord, 2001). The contrasting stable carbon-isotope patterns between the west and the east show that this lateral climatic contrast has been in place since ~7 Ma (Vögeli et al., in review). Lupker et al. (2012) showed that sediments are predominantly weathered in the floodplain; rapid transport through the floodplain therefore inhibits extensive weathering. The apparent contradiction between a more humid climate and less intense weathering in the east can be resolved by hypothesizing that sediment storage in the floodplain was less important, due to higher runoff and more efficient sediment transport. A similar contrast can be observed when comparing the marine records of the Indus and the Bengal Fans (Clift et al., 2008).

The Indian Summer Monsoon is the main driver for the precipitation pattern along the Himalayan front. The most recent studies on the onset of the Indian Summer Monsoon suggest that it was active in Eocene times (Licht et al., 2014). We can therefore assume that monsoonal winds transported moisture from the Bay of Bengal to the Himalaya since this time, although the variation in monsoon strength over time remains to be evaluated. Marine records of the Bengal and the Indus fans have been used to reconstruct paleoclimate in the Himalayan region (Clift and Gaedicke, 2002; Clift et al., 2008; France-Lanord et al., 1993, amongst others). Figure 8 shows K/Si vs Al/Si ratios for sedimentary rocks of the western and eastern Siwaliks together with modern sediments of the Ganges River (Lupker et al., 2013), and sediments from the Indus fan (~16-1 Ma; Clift et al., 2008). The overall westward increase in weathering intensity that we infer from our Siwalik sections is confirmed by including these additional data, as Indus-fan sediments appear to be slightly more weathered than both the proximal Siwalik record and the modern sediments of the Ganges River. Thus, sediments in the drier, more seasonal western Himalaya seem to be generally more weathered than in the more humid but less seasonal eastern part of the belt, and have been so since the Miocene.

6. Conclusions

The Himalayan foreland basin is an excellent laboratory to study lateral variations in the evolution of climate and weathering regimes, as foreland-basin sediments of the Siwalik Group crop out along the entire mountain front and contain a continuous record of tectonics, erosion, climate and weathering. The compilation of three sections (JN, JW, HK) in the western Himalaya allows reconstruction of climate and weathering as far back as the Early Miocene. The lack of longer records in the east limits direct comparison between the western and eastern Himalayan foreland to mid-Miocene and later times. By studying clay mineralogy and major-element compositions, taking potential provenance and diagenetic biases into account, important insights into past weathering and climatic regimes can be gained. The K/Al ratio in particular appears to be a robust proxy for past weathering intensities.

The smectite content of Himalayan foreland-basin sediments increases from 11 Ma in the west and 8 Ma in the east, respectively. Illite crystallinity does not increase down-section, suggesting that illitisation of smectite did not occur in the studied sections. The influence of diagenesis can therefore be considered negligible and the change in clay mineralogy can be ascribed to a change towards a more seasonal climate. The relative abundance of kaolinite over smectite in the east is consistent with a more humid but less seasonal climate. K/Al is also generally higher in the east, indicating less intense weathering. Less intense weathering in the eastern Himalaya is interpreted as resulting from a more humid but less seasonal climate, leading to more erosion and runoff. The resulting shorter residence time of sediments in the floodplain can explain the observed less intense weathering. In both the west and the east, a slight trend towards more intense weathering with time can be observed, which we interpret as being due to a climatic evolution toward more seasonality. Throughout the studied time span, seasonality appears to be stronger in the west of the mountain belt than in the east, as it is today. During the period between ~7 and 5 Ma, sediments in the west are particularly unweathered; however, we link this excursion to a change in provenance and depositional environment, rather than a major change in climate.

Reconstruction of past climate and weathering regimes remains a challenging task. The monsoonal climate and its evolution is still not fully understood. In particular, the monsoonal influence on seasonality linked with overall humidity needs to be further investigated. Paleo-vegetation, clay mineralogy and major elements can be used as indicators for seasonality, but more such paleo-seasonality studies are crucial to understand the evolution of the monsoon.,

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Figure captions

Figure 1: Geological map of the Himalaya, after Hirschmiller (2014). Red lines indicate the Siwalik sections analyzed in this study: JN: Joginder Nagar, JW: Jawalamukhi, HK: Haripur Kolar, and KM: Kameng. Other abbreviations: MFT: Main Frontal Thrust; MBT: Main Boundary Thrust; MCT: Main Central Thrust; STD: South Tibetan Detachment.

Figure 2: Stratigraphy, sampling points, clay-mineral assemblages and inferred provenance of the Joginder Nagar (JN), Jawalamukhi (JW) and Haripur Kolar (HK) sections. Stratigraphy and age dating after Meigs et al. (1995), Sangode et al. (1996) and White et al. (2001). Provenance from White et al. (2002), Najman et al. (2009) and Suresh et al. (2004).

Figure 3: Comparison of clay-mineral assemblages and illite crystallinity from the western (empty circles) and eastern (black diamonds) Himalayan samples. Eastern Himalayan samples represented in grey diamonds show anomalous clay-mineral assemblages, due to very low clay concentration (4 Ma) and proximity to a local thrust (13 Ma) (cf. Vögeli et al., accepted).

Figure 4: Correlation between SiO_2, K_2O and Al_2O_3; compilation of all samples of the western sections (JN, JW, HK). Mineralogy and major-element concentrations vary from fine- to coarse-grained sediments; fine-grained sediments (in black) show relative depletion in SiO_2 and enrichment in K_2O and Al_2O_3.

Figure 5: Variation of K/Al and K/Si* ratios with time. Red dots represent the western Himalaya (JN: Joginder Nagar; JW: Jawalamukhi; HK: Haripur Kolar sections) and blue dots the eastern Himalaya (Kameng river section; KM). Lighter red and blue dots are coarse-grained samples; darker dots are fine-grained samples. Samples in the west generally show a higher degree of weathering except between 7-5 Ma, when weathering intensity appears exceptonally low in the west. Red and blue lines are eight-point moving averages showing general trends of weathering over time.

Figure 6: CaO vs. SiO_2 concentrations. Red circles show western sections; blue circles are sediments of the Kameng River section in the east. Calcium concentration is elevated in the western Siwaliks, but does not correlate with SiO_2, hence is not dependent on grain size.

Figure 7: Evolution of the K/Si ratio of fine- and coarse-grained sediments of the western (black) and eastern (grey) Siwaliks. Dashed lines represent the 0.95 confidence level of the linear regressions of the west and east (method after Lupker et al., 2013). Anomalous values between 8 and 5 Ma are encircled in orange, orange line shows the regression excluding these anomalous points.

Figure 8: Comparison of K/Si vs. Al/Si ratios of the western and eastern Siwalik sediments with those of marine sediments from the Indus Fan (Clift et al., 2008) and modern sediments of the Ganges River (Lupker et al., 2013).

Appendices

Appendix 1: H2O+/Si* ratio vs age

Appendix 2: XRD diffractograms of clays of similar age from west (red line) and east (blue line), showing variations in the evolution of the clay mineralogy between west and east; smectites are similar in the west and the east.

Table S1: Whole-rock major-element chemistry analyses of samples from the Joginder Nagar, Jawalamukhi, and Haripur Kolar sections, Himachal Pradesh, northwestern India.