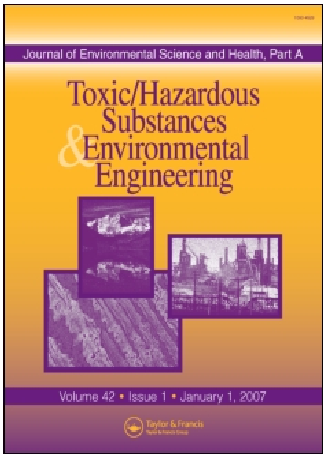

Journal of Environmental Science and Health, Part A
Toxic/Hazardous Substances and Environmental Engineering
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713597268

Bengal arsenic, an archive of Himalaya orogeny and paleohydrology

Stephane Guillot [a]; Laurent Charlet [a]
[a] University of Grenoble, Grenoble, cedex 9, France

Online Publication Date: 01 October 2007
To cite this Article: Guillot, Stephane and Charlet, Laurent (2007) 'Bengal arsenic, an archive of Himalaya orogeny and paleohydrology', Journal of Environmental Science and Health, Part A, 42:12, 1785 - 1794
To link to this article: DOI: 10.1080/10934520701566702
URL: http://dx.doi.org/10.1080/10934520701566702

Journal of Environmental Science and Health Part A (2007) **42**, 1785–1794

ISSN: 1093-4529 (Print); 1532-4117 (Online)
DOI: 10.1080/10934520701566702

Taylor & Francis
Taylor & Francis Group

Bengal arsenic, an archive of Himalaya orogeny and paleohydrology

STEPHANE GUILLOT and LAURENT CHARLET

University of Grenoble, OSUG – CNRS, 1381 rue de la Piscine 38041 Grenoble cedex 9, France

Holocene groundwater in many districts of the West Bengal and parts of Bangladesh are enriched in arsenic enhancing poisoning effect on humans. One of the main problems to depict the source of arsenic is that this element is very mobile and can be easily removed and recombined from the source during alteration processes, transport and mobilization in sediments. The Ganga-Brahmaputra river system mainly contributed to the buildup of the Bengal fan, which is considered one of the largest modern deltas of the world, then the possible source of the As has probably to be search within the Himalayan belt. We propose that the Indus-Tsangpo suture zone dominated by arc-related rocks and more particularly by large volume of serpentinites enriched in arsenic could be one of the primary source of arsenic. The fact that, the present day arsenic concentration in the main Himalayan river, and particularly the Siang-Brahmaputra river system is not so high as expected can be explained by strong aridic conditions present day prevailing in the Indus-Suture zone and do not favored the weathering of serpentinites into As rich-smectite and Fe-hydroxydes. For the Ganga basin, the original source of arsenic has to be search in the weathering of arc related rocks in the Indus-Tsangpo suture zone followed by its intermediate storage into the sediments of the Siwalik foreland basin, playing the role of arsenic reservoir from Miocene to Pleistocene. Intense tectonic activity in the front of the Himalayan belt associated with high rainfall conditions during the Holocene allowed the arsenic to be remobilized and transported toward the Bay of Bengal.

Keywords: Arsenic source, Serpentinite, Indus-Tsangpo, Siwalik Himalaya, Bay of Bengal; Indus-Tsangpo.

Introduction

It is well known that groundwater in many districts of the West Bengal and parts of Bangladesh are enriched in As enhancing a poisoning effect on humans.[1−7] The As content in groundwater in the Bengal delta varies from 0.05 to 3.7 mg/l with an average of 0.2 mg/l, which is 20 times higher than the limit recommended by WHO in drinking water.[7] Although the cartographic distribution of As concentration in Ganga-Bramaputra, Mekong and Red River fans are now relatively well modified, the primary source and its links with the paleohydrology remain unclear. One of the main problem in depicting the source of As is that this element is very mobile and can be easily removed from the source during alteration processes, then transported in solution in water, or in suspended particles, adsorbed onto various phases such as micas or Mn-phases, or recombined with sulfides of Fe-oxyhydroxides, before being scavenged in reducing conditions in groundwater.[5−10]

Address correspondence to Stéphane Guillot, Maison des Maison des Géosciences, University of Grenoble, 1381 rue de la Piscine. 38041 Grenoble Cedex 9, France. E-mail: sguillot@ujf-grenoble.fr

It has been suggested that the primary geogenic sources of the As in the groundwater in the Bengal basin are As-rich metal deposits in the Ganges basin and in the Darjeeling Himalayas, As-bearing pyrite coming from the Gondwana coal seams and from the Rajmahal volcanics.[1,5,6,11,12] However, as emphasized by Stüben *et al.*[7] except for a few horizons with higher values of about 100 to 200 ppm the bulk As content of the aquifer sediment is comprised between 1 to 30 ppm[4,5] i.e., the normal range of the As content of sedimentary rocks.[13] Therefore a diffuse source of the origin of the As has been proposed rather than a precise source.

The Ganga-Brahmaputra river system mainly contributed to the buildup of the Bengal fan, which is considered one of the largest modern deltas of the world.[14,15] Ever since the Tertiary, this river system has carried enormous volumes of sediments from the Himalayan belt. Consequently the possible source of the As has probably to be search within the Himalayan belt. It is noticeable that the average annual sediment transports by the Ganga-Brahmaputra river system is of about 1800 tons/km^2, with suspended load estimated between 540 to 1157 millions tons/yr.[14] It is the large-river basin with the highest denudation rate on Earth (0.69 mm/yr according to Summerfield and Hulton.[1−7] We propose in this paper to investigate

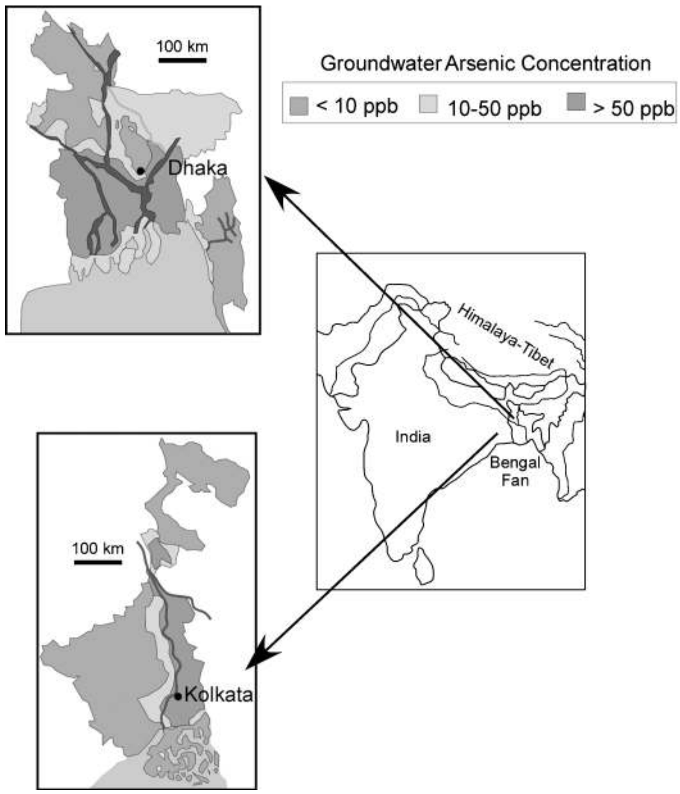

Fig. 1. Arsenic distribution in the Ganga-Brahmaputra basins.[3]

the petro-tectonic assemblages of the Himalayan belt and its hydrologic system through time in order to propose potential sources for As as guideline for further investigations.

The Bengal Fan system

In terms of the numbers of people at risk, the high-arsenic groundwaters in the alluvial and deltaic aquifers of Bangladesh and West Bengal represent the most serious threat to public health. Groundwaters survey indicated that than 60% of the wells in South Bangladesh and West Bengal contain more arsenic than the national standard fixed at 10 $\mu g \cdot L^{-1}$[17] (Fig. 1). The aquifers are generally shallow (<100 meters deep) and consists of Holocene micaceous sands, silts and clays associated with the Ganga, Brahmaputra and Meghna rivers (Fig. 2a). Positive correlation between arsenic and iron in the groundwaters has been reported at the local scale.[18]

It is noticeable that the alluvial and deltaic sediments have a total arsenic content of 2 to 20 ppm; these values are close to world-average concentrations for such sediments. Moreover, deeper groundwaters, in older Plio-Pleistocene sediments (2.5 to 1.5 Ma) in northern and western Bangladesh have arsenic concentrations below 5 $mg.L^{-1}$[17] (Fig. 2b). These two observations suggest that deeper and older sediments are subject to longer periods of groundwater flows aided by greater hydraulic heads during glacial periods. These sediments will, therefore, have undergone a greater degree of flushing and removal of labile solutes than Holocene sediments (10,000 years BP) at shallower depths.

The BGS studies reveals the following average concentration of arsenic (mg/kg or ppm) in the bedload of the main rivers of the Bay of Bengal: Ganga river: 2.03 ppm, Brahmaputra: 2.76 ppm; Meghna river: 3.49 ppm. In the Meghna Estuary, 90% of the total arsenic is bound to nearly insoluble phases of the suspended matter (e.g., crystalline iron oxides, residual minerals such as micas, fedspar and chlorite). Arsenic is enriched in the suspended matter by a factor of 2 relative to the sediment composition, but the arsenic enrichment does not follow the concentration of Mn and other transition metals (Zn, Ni, Cr, Co).

It is more similar to the concentration of iron. [20] These authors also show that the sediments from the Bay of Bengal are dominated by quartz (40–55%), plagioclase (14–21%), alkali feldspar (2–10%), muscovite (illite) (2–7%), clay minerals (chlorite-illite-kaolinite) representing 4 to 10% and heavy minerals (zircon, ilmenite, rutile, amphibole and garnet). Thus, metamorphic rocks and intermediate-to-acid magmatic rocks are the original source of the sediments. The Brahmaputra-Ganga river system is the big-river basin with the highest denudation rates on Earth.[16] Present erosion is very low in two-thirds of the basin, including the elevated Tibetan plateau, the Assam region, the Bangladesh and the Indo-Gangetic plain (Fig. 3).

The highest erosion rate varies strongly in the Greater Himalayan belt and reach peaks in the eastern syntaxis.[15] The Siang river, draining the eastern syntaxis contributes of around 25% of total sediments reaching the Bay of Bengal, whereas the Ganga basin, which is four time large, contributes to 40% of the total sediment of the Bay of Bengal. The rest comes from Himalayan tributaries (14%) and 7% from the Shillong plateau.[15] As most of the Himalayan units are dominated by crystalline rocks and sediments usually poor in arsenic (<2 ppm[17]), only two zones are potentially arsenic-rich, the Indus-Tsangpo suture zone due to the abundance of arc related rocks usually rich in arsenic.[21] and the Siwalik Neogene sediments due to the abundance of clays minerals also enriched in arsenic.[22] Next, we investigate the Himalayan geology and its links with As anomaly.

The Himalayan belt

The Himalaya rises abruptly from the Indo-Gangetic plain to high mountain peaks, south of the Indus-Tsangpo suture zone all 2500 km long (Fig. 3). The main tectonic units can be followed continuously from south to north. The north-dipping Main Frontal Thrust (MFT), active since ~2 Ma, places the Siwalik Group over the present-day Indo-Gangetic foreland basin. The Main Boundary Thrust, active since at least 10 Ma[23] places the Lesser Himalaya

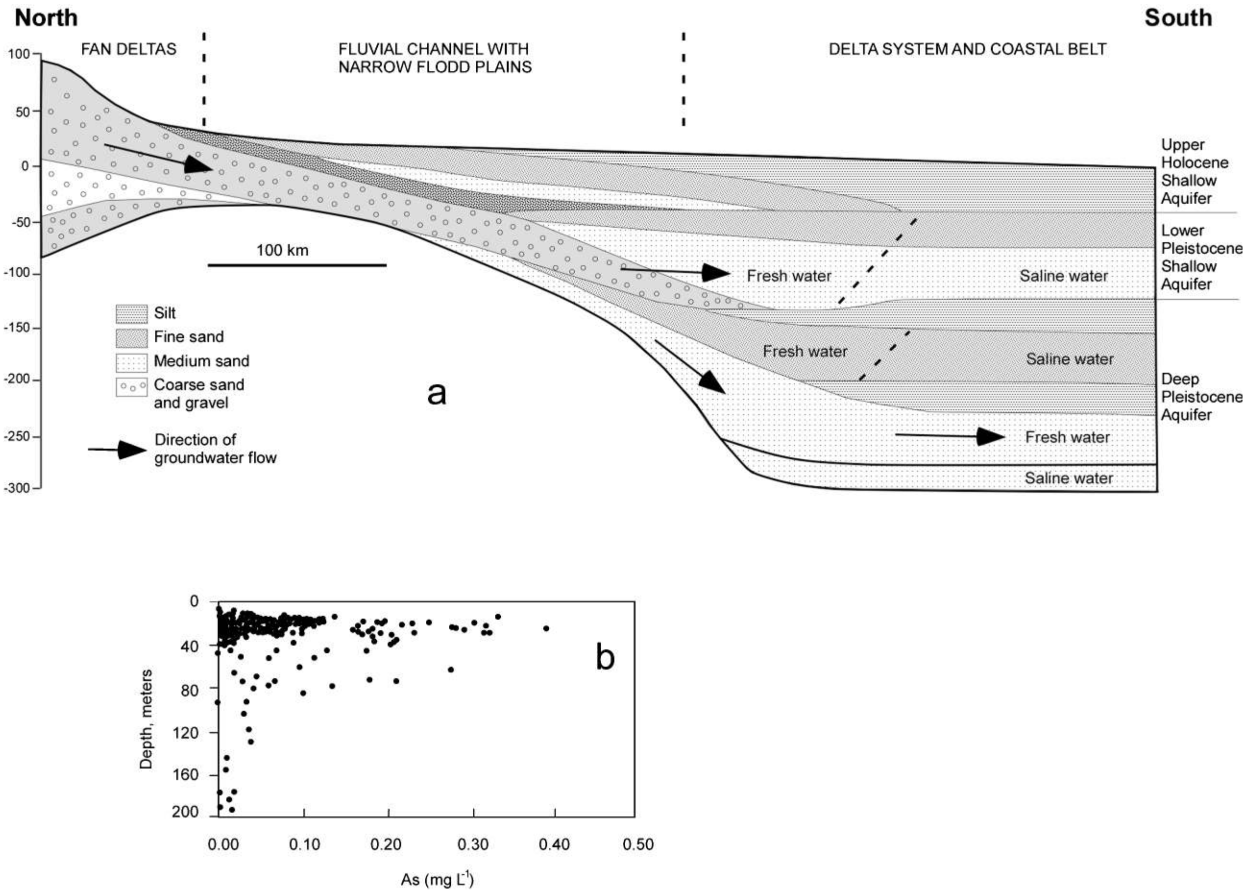

Fig. 2. (a) Hydrogeological cross section across Bangladesh. The upper shallow aquifer located in the flood plains are located in the Holocene sediments. They are characterized by fine sands cover silts and clays, impermeable to rainwaters.[19] (b) Distribution of As concentration in groundwaters as a function of screen depth in the Chakdaha block, west Bengal.[18]

Proterozoic metasediments over the Sub-Himalaya molasse of the 4 km thick Siwalik Group. Above, the South Tibetan Detachment system separates the Higher Himalayan Crystallines (HHC) from the Paleozoic to Eocene sediments of the Tethys Himalaya. Northwards, the basement of the Tethys Himalaya outcrops as domes, the northern Himalayan massifs. Finally, the Indus Tsangpo suture zone (Fig. 3), squeezed between the Tethys Himalaya and the Asian plate, includes Indian continental slope and rise sediments, slices of Cretaceous ophiolite mélange and Cretaceous to Paleogene forearc basin sediments.[24]

The Siwaliks

Potential adsorption substrates and co-precipitation host minerals are abundant throughout the fine-grained Siwalik levels, as iron mineralization, sulphides or as clays dominated by smectite.[25] The Siwalik Groups defines the synorogenic continental sediments of the foreland basin.[23,26] They are located beneath the Gangetic plain (Terai plain in Nepal) or in the outer part of the fold and thrust belt.[27,28] (Fig. 3). The base of the Siwalik Group is dated between 15.5 and 13 Ma in India and Nepal.[26,29] The Siwalik Group represents a typical fluvial fining upward succession, with lower unit consisting of fluvial channel sandstone and oxidized calcareous paleosols, the middle unit is thick channel sandstones and the upper unit comprised mainly gravely braided river deposits.[28]

Most of the sandstone has a variable silty clayed matrix and some of them should be called graywacke. The clay mineral assemblage was observed in the <2-μm fraction and are dominated by the illite, chlorite, kaolinite, smectites and mixed layers (illite-smectite/chlorite-smectite). Illite and chlorite rich assemblage dominated before 9.5 Ma. Between 9.5 and 6.5 Ma, smectite/chlorite alternate with illite/chlorite assemblages. After 6.5 Ma, smectite and kaolinite assemblages dominate.[28]

According to Nd isotopic data, Huygue *et al.*[28] estimated that before 10 Ma, the Lesser Himalaya represents less than 10% of sediment input and increased up to 30% after 10 Ma. It has been proposed that the formation of smectites after 10 Ma results from feldspar weathering;[30] After 6.6 Ma, one can not invoque smectite related to feldspar weathering as the observed feldspar content in these deposits is lower than during the 6.5 to 9.5 Ma period.[28] These authors suggest that from 6.5 Ma onward,

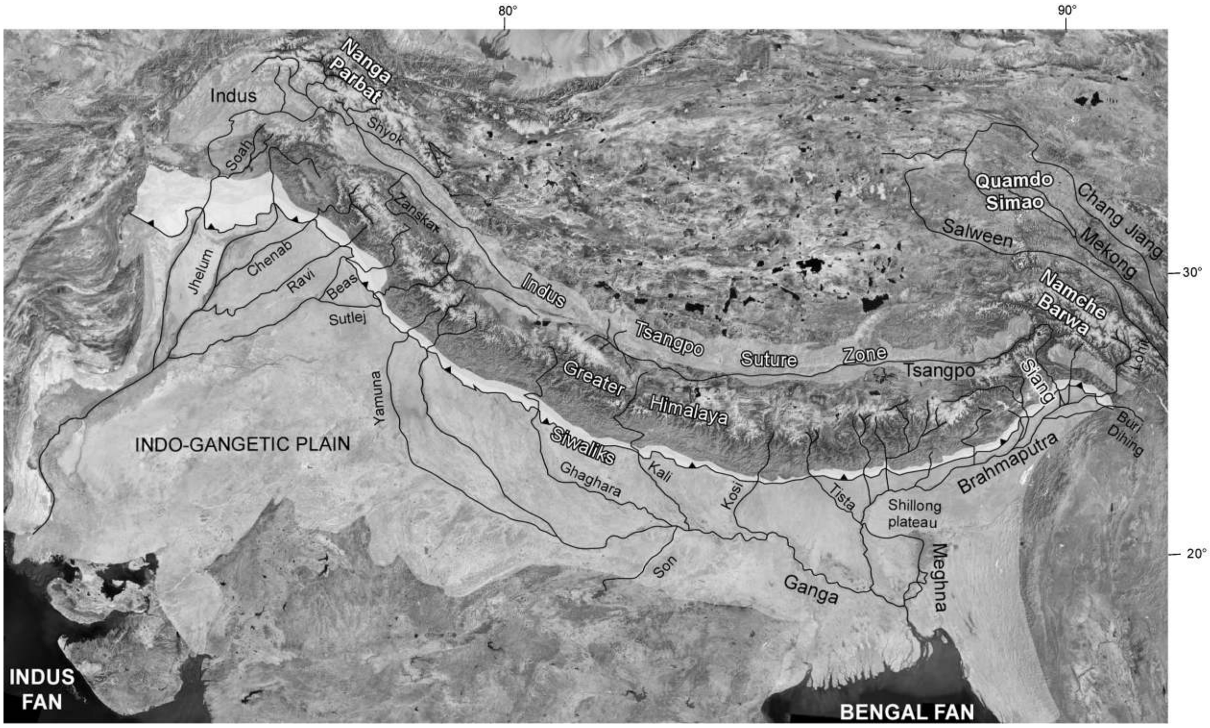

Fig. 3. Himalayan orographic map with the major drainage systems and the main Himalayan geologic zones.

the smectite apparition is linked to strong seasonality that favored its formation from pedogenetic processes. An alternative is that this smectite assemblage comes from the direct weathering of serpentinites.[31]

Several north-dipping thrusts delineate tectonic boundaries in the Siwalik Group.[32] These thrusts branch off the main decollement, which lies at a depth of 4–5 km.[27] The most frontal one is the MFT and is activation since 2 Ma is reponsible to the uplift and consequently erosion of the Siwalik sedimentary basin.[32] The shortening that occurs along the MFT is not constant through time and the proportion that occurs along the MFT can be estimated by the ratio of MFT shortening rate to total Himalatan shortening rate estimated at 20 mm/yr since the Miocene.[27,32,33] Mugnier *et al.*[32] estimated that during the Plio-Pleistocene, only 20% of the motion occurred on the MFT and since the Holocene, the MFT concentrated between 50 and 100% of the whole motion.

This suggests a strong activation of the MFT during the last 10,000 years, enhancing an important uplift and erosion of the Siwalik. The exposed Siwalik form a 10 to 50 km wide and some 1800 km long. As it is composed of immature argillaceous and arenaceous sediments, they erode rapidly to a yield high fraction of the modern fluvial sediment. Chesley *et al.*[34] shown that in the Siwalik, paleosols form by weathering of floodplain sediment by local rainwater, downward leaching of dissolved weathering products, and precipitation of secondary carbonates nodules, Fe/Mn oxides and surrounding Fe-oxide matrix that sorbed the arsenic. The same fluids pass through into local rivers and the weathering of floddplain sediments determines the elemental compositions of local rivers going to the Ganga-Brahmaputra rivers.[35]

Recent studies of arsenic groundwater in the Terai plain of Nepal.[22,36,37] and in Pakistan and India,[6,38] proposed that high-arsenic groundwaters are related to the eroded Siwalik. In fact, sediments carried from the Siwalik by the minor rivers release more arsenic than those carried by major rivers from the Higher Himalaya.[22] Analysis of fan-watershed pairs, covering all of Nepal, indicates correlation of both average and maximum arsenic concentration in shallow groundwater from alluvial fans to the proportion of range-front Plio-Pleistocene Siwalik source rocks in the erosional part of watersheds, where fan sediment is produced. Areas of greatest arsenic contamination occur on fans where Siwalik rocks are the only sediment source, although areas free of arsenic contamination also occur on the same fans. On fans lacking Siwalik sediment, arsenic contamination is rare, despite the presence of secondary ferric oxyhydroxides and detrital pyrite containing arsenic.

Van Williams[37] already proposed to explain this relationship that much of the arsenic in groundwater came from oxidation of sulfides in the alluvial aquifer and authigenic sulfides found in Siwalik debris break down more readily than primary hydrothermal sulphide minerals found in debris of veins eroded from metamorphic rocks of the middle and Higher Himalaya. In contrast, Gurung *et al.*[22] proposed that large-scale difference in the sediment source can explain local variation of arsenic concentration observed in the Terai plain.

Eastward, the main tributaries of the Brahmaputra river (Fig. 3) cross-cut and eroded the Siwalik Group; according to Stanger[40] the Siwalik is the precursor source of arsenic analysed in the Brahmaputra. If the Siwalik Group is the probable reservoir of the arsenic transport by the Ganga river system towards the West Bengal Bay, it cannot be the original source, as it corresponds to detrital foreland deposit. He[40] proposed that the Quamdo-Simao volcanic and ophiolite province located north of the Namche Barwa syntaxis (Fig. 3) is the original source of arsenic transported during Miocene time toward the Siwalik foreland basin. Next, we will demonstrate that the Quamdo-Simao volcanic and ophiolite province cannot be the source for arsenic observed in the Siwalik and in Himalayan flooding plain as the main rivers coming from this ophiolitic province never flowed in this direction. In contrast, the Indus-Tsangpo suture zone that presents the same lithological characterics as the Quamdo-Simao volcanic and ophiolite province is the potential source for arsenic, feeding the Siwalik Group during Miocene and Pleistocene time, before to be removed dring the Holocene.

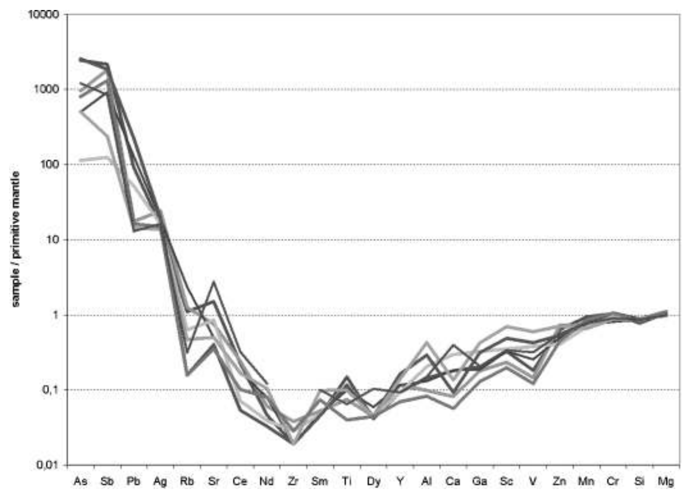

Fig. 4. Composition of serpentinites from the Indus-Tsangpo suture zone (normalized to primitive mantle). As and Sb are similar in compatibility to light rare earth elements, but they are place on left side of the diagram to emphasize their fluid-soluble enrichment (after Hattori and Guillot[45]). Notice the strong enrichment in As compared to the primitive mantle and related to As rich fluid released during the subdution processes.[45,46]

The Indus-Tsangpo suture zone

The Indus-Tsangpo suture zone marks where the Tethys Ocean was consume as India approached and ultimately collided with Asia, 55 Ma ago.[24] The most widely accepted tectonic model for this event is one in which the entire N-S extent of the Tethyan oceanic lithosphere was subducted along the southern margin of the Lhasa-Karakorum block defining an Andean-type margin from 110 to 50 Ma, dominated by calc-alkaline plutonic rocks (mainly diorite and granodiorite with subordinate andesitic to rhyolitic volcanics), and along an intra-oceanic subduction system from 120 to 70 Ma.[41]

The particularity of the intra-oceanic paleo-subduction zone is that a large part has been obducted, defining a succession of large dismembered ophiolitic units with associated ophiolitic melange from the Kohistan arc in the west to the Namche Barwa syntaxis in the east (Fig. 3).[40−43] Their crustal section, generally 1 to 5 km thick is marked by the abundance of MORB type volcanics (pillow lava and dyke swarm) overlying a mantle section dominated by more or less serpentinized peridotites. The peridotites are dominated by the abundance of harzburgite and dunite, typical of the arc environment.

Locally strongly serpentinized depleted peridotites are associated with eclogitic rocks, this is the case in the eastern Ladakh;[44] but also close to the Nanga Parbat syntaxis in Pakistan.[45] The serpentinites consist mainly of antigorite. Sulfides minerals are largely absent except for minute grains of millerite (NiS) and heazlewoodite (Ni_3S_2). All serpentinites have similar bulk chemical compositions with high concentrations of compatible elements: high Cr ($\sim$2000 ppm), Ni ($>$2000 ppm) and MgO ($>$40 wt%). Overall high contents of platinum elements and Re bulk samples with high Cr ratio in spinel are consistent with the refractory mantle origin of the serpentinites.[44] However the serpentinites are enriched in solubles elements such as As, Sb or Pb. Arsenic content reaches up to 275 ppm (Fig. 4).[45] Electron microprobe study on Himalayan serpentinites identified minute sulfide minerals with high As in heazlwoodite (Ni_3S_3) and tinity grain of arsenide. Most magnetites contain low As content below 16 ppm, in contrast large antigorite grains yielded varying As values up to 90 ppm.[46]

XANES spectra of Himalayan serpentinites do not show evidence for a contribution of As from sulfide minerals. Hattori *et al.*[46] proposed that the arsenic in the serpentinites from the Indus suture zone is mainly concentrated in the antigorite by substution of Si(IV) by As(V). Arsenic may be also partly sorbed on the magnetite surface. It has been proposed that the arsenic-rich concentration in the serpentinites involves the release of As(V) from subducted sediments to the mantle wedge beneath the volcanic arc.[45] These serpentinites as the base of the arc acted as a sink for water and arsenic and then exposed to the surface by collision processes.

It is noticeable that the Indus Tsangpo suture zone is exposed to the surface for a long time as zircon and apatite fission tracks indicate that the internal zone was near the surface since 40 Ma.[47] Thus, they have been exposed to weathering for a long time. Weathering processes of serpentinites result to a leaching of Mg and Si and relative enrichment in the least mobile elements, i.e., Fe and Al producing smectite, kaolinite, chlorite and large amount of oxyhydroxides minerals.[31] It is noticeable that oxyhydroxides can contain up to 76,000 ppm of arsenic, Pichler *et al.*[48]

emphazing that weathering of serpentinites is probably one of the most important source of arsenic.

Himalayan paleohydrology

Four main stages in the collision of India with Asia can be recognized.[24] The first stage from Late Cretaceous (~100 Ma to 55 Ma) involves the accretion of oceanic arcs onto the northen Indian margin and the southern Asian margin and ended with final closure of the ocean Tethyan domain as the Indian margin subducted beneath the Asian-Adean type arc. The second stage from 50 to 25 Ma involve the progressive underthrusting of the Indian plate beneath south Tibet accompanied by progressive thickening of the future Tibetan plateau as convergent deformation progressed from the initial southern collision site to the northern edge of the Tibetan plateau.[49] The third stage, from about 25 Ma to 5 Ma involves transfer of the main shortening to areas south (Himalaya) and north (Tien Shan) of the Tibetan plateau. This phase was accompanied by lateral extrusion of Indochina with a probable uplift of the Tibetan plateau.[49] The fourth stage, from about 5 Ma to the present is characterized by the activation of the Main Frontal Thrust in Himalaya and widespread extensional tectonics in south Tibet.

Several authors attempted to restore the evolution of the main drainage systems during the collision of India with Asia. White *et al.*[50] suggest that the Greater Himalaya clast (poor in As) first appeared in the Himalayan foreland basin at the end of the Oligocene (~25 Ma) and high-grade metamorphic minerals (kyanite and sillimanite) did not appear until deposition of the Middle and Upper Siwalik that are younger than 11 Ma. Yin[51] reinterpreted the data of White *et al.*[50] proposing that the 5 to 30 Ma Ar-Ar detrital mica ages obtained from the NW Foreland basin do not reflect the ealier exposition of the Greater Himalaya, but come from the erosion of the Internal Crystalline massif close to the Indus suture zone (rich in As) and probably at the surface since a minimum of 20–30 Ma.[47] Finally, Yin[51] proposed that in western Himalaya, the Greater Himalaya was not exposed prior to 10 Ma. In central Himalaya, in Nepal, existing Ar-Ar muscovite ages show that the Greater Himalaya was at a depth of about 10 km between 15 and 13 Ma.[52,53]

This implies that the Greater Himalaya could not be exposed prior to 15 Ma–13 Ma, this is an important constrain for the potential sources areas for Arsenic. Moreover, high-grade metamorphic minerals indicative of the Greater Himalaya did not appear in the central Himalayan foreland basin until after 11 Ma or even later during the deposition of the Lower Siwalik Group.[54] This confirms that the Greater Himalaya did not reach the surface until after 11 Ma.[51] This strongly contrasts to the long-held view that the Greater Himalaya had already exposed at the surface since a minimum of 17 Ma.[55] and suggest that the Indus-Tsangpo suture zone rich in As minerals feed the Siwalik foreland basin during the Miocene.

A possible history of the relationships between the hydrogeology pattern, the erosion cycle and the deposition can be proposed (Fig. 5). The Paleogene Himalayan foreland basin receives rich sediments mostly from southern Tibet and the Indus-Tsangpo suture zone. These sediments result of the mechanical and chemical alteration of calc-alkaline magmatic rocks, basic and ultrabasic rocks enriched in arsenic and of the UHP metamorphic units in northwestern Himalaya. In absence of reliefs to the south, the main rivers flowed directly towards the south.

In the Early Miocene (after 25 Ma), the foreland basin receives sediments from the Tethyan Himalaya and until after 11–5 Ma, the Greater Himalaya already exposed started to be eroded and high-grade metamorphic clasts poor in As, first appear in the Siwalik Group of the Himalayan foreland (Fig. 5a). According to Yin[51] the rivers that feed the Siwalik and Bengal bay up to 5 Ma come from the Indus Tsangpo suture zone and consequently a part of the sediments comes from As-rich sources. Mugnier and Huygue[58] proposed that breakoff of the Indian slab during the Miocene increased the altitude of the internal Himalayan belt and consequently the erosion.

France-Lanord *et al.*[55] and Huygue *et al.*[28] shown that during this period the Greater Himalaya was extensively exposed, leading to deposition of large amount of metamorphic clasts in the Siwalik Group and the Bay of Bengal. Even though the Himalaya had become a mountain of prominence by mid Miocene, it could not have been an orographic barrier high enough then to prevent the movements of heavy-bodied bulky quadrupeds like the hippopotamus, rhinoceros and elephant in and across the lake basins of Potwar, Jammu, Kashmir and Kathmandu. There was indeed a youthful mountain characterized by mild relief and gentle topography.[59]

The period of tectonic quiescence was broken by a powerful tectonic upheaval in the temporal interval 11 to 7.5 Ma (Fig. 5b), when the gentle highland became a mountain barrier. Revival of movements on faults that delimit the boundaries of four terranes of the Himalayan province were responsible for spectacular uplift of the Himalaya approximately 8 Ma ago.[60] These tectonic events of the late Miocene were accompanied by sudden and drastic change in the climate conditions. The old geomorphically mature topography was rejuvenated, and the landscape underwent reshaping. When heavy seasonal rainfalls started beating the newly emerged mountain, great volumes of detritus eroded from the uplifting terranes found their ways to the foreland basin and to the Indian Ocean. Denudation of the Himalayan province had begun in mid-Miocene as evident from the influx of clastic sediments associated with deep-water microfauna in the Bay of Bengal and Andaman-Nicobar domain.[61]

The major change occurred after 5 Ma (Fig. 5c), this period corresponds to a strong reorganization of the

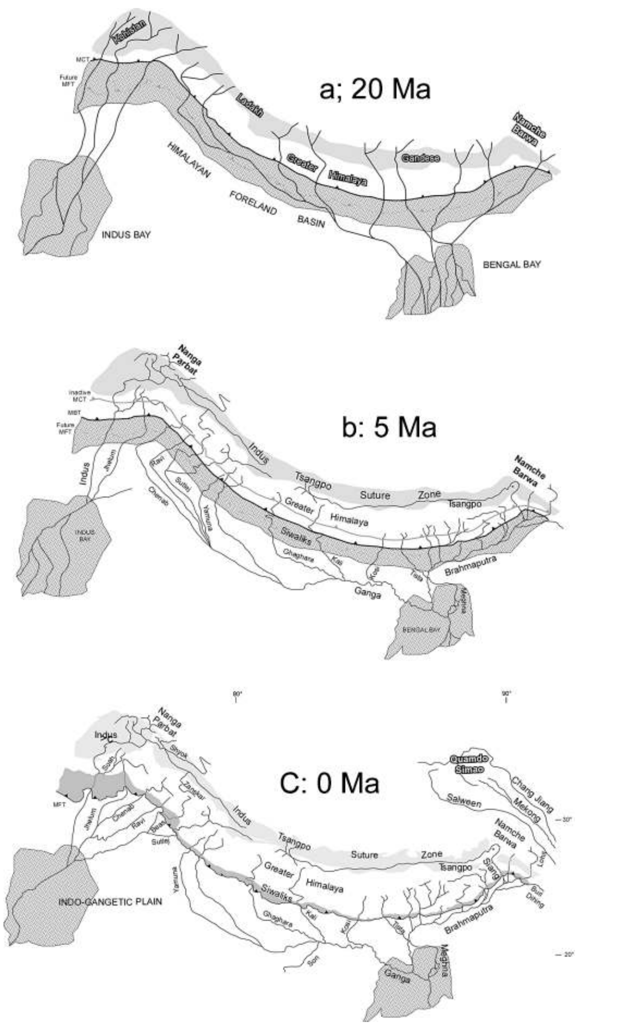

Fig. 5. Paleohydrology drainage system of the Himalayan belt and depositional location of the Himalayan foreland basin through time (modified after Brookfield.[58] Najman *et al.*[26] and Yin[51]). (a) During the Miocene (20–10 Ma), the Main central thrust (MCT) was active leading to the development of a large foreland basin, but the Himalayn barrier did not exist and the main rivers probably flowed directly from the South Tibetan plateau to the foreland basin leading to important weathering of the Indus-Tsangpo suture zone, transport of As and its immobilization in the Siwalik foreland basin. (b) During the upper Miocene-Pleistocene (10–5 Ma), the MCT was inactive whereas the Main Boundray thrust leading to the exhumation of the Lesser Himalaya and the deposit of the upper Siwalik Group. The Himalayan barrier was probably present. In the Western syntaxis, the Indus river flowed directly toward the Indus bay, whereas the Tsangpo river is progressively captured by the Siang-Brahmaputra river system. In the central part of the belt, most of rivers flowed directly from the Greater Himalaya towards the Ganga drainage system in the direction of the Bay of Bengal. (c) after 5 Ma to present, most of the NW Himalayan rivers are captured by the Indus river system river whereas the Siwalik was strongly uplifted and eroded, due to the activation of the MFT. The combination of strong tectonic activity and a more humid climate during the the Holocene leads to the removal of As stored in the Siwalik sediments and its transportation by the Ganga-Brahmaputra rive-system towards the Bay of Bengal.

Himalayan river.[58,62] In the Indo-Gangetic plain, the major Punjab rivers (Chenab, Ravi; Beas; Sutlej) which flowed east into the Ganga river before that time are derouting into the Indus[62] whereas in the eastern syntaxis, the Tsangpo-Bramaputra river is capture by the Brahmaputra river due to the rapid uplift the Nanga Parbat syntaxis.[58] Moreover, the initiation of MFT located front of the Siwalik Group took place after 2 Ma with an important recent activity (shortening around 20 mm/yr from 0.3 to 0.1 Ma) leading to an important incision and erosion of the uplifted Siwalik Group.[32]

The Holocene period alternate between high rainfall and aridity favouring weathering of Himalayan rocks. The prolonged period of late Quaternary dryness, including the time of Last Glacial Maximum (20,000 to 16,000 yr BP) persisted until nearly 11,000 yr BP. Then the climate became progressively wetter and warmer as the SW monsoon intensified, reaching its peak in the early to middle Holocene. The wet and warm epoch was followed by a time of severe aridity around 3500 yr BP and lasting until about 2000 yr BP.[59] In central Indo-Gangetic plains heavy rainfall washed away salts and carbonates from the soil and caused development of a better drainage around 8000 yr BP.[63] Pollens from the Dokrani peat in the Gangotri glacier area in the Greater Himalaya (in Garhwal) indicate a warm–wet spell of climate between 6500 and 4000 yr BP.[64] To the far northwest in Ladakh, where serpentinites are abundant, pollens of the Tsokar lake testify to the expansion of flora under warmer–moist condition around 10,000 yr BP.[65] Along the Indus-Tangpo suture, the water level of the Bangong Co lake reached the maximum about 6000 yr BP.[66] testimony to higher rainfall in the period 7500 to 3000 yr BP.

Tracing Arsenic in Himalaya

As discused above, the As path from the river to the groundwater in the Bengal Bay is now relatively well constrained. The hypothesis that As is fixed on Fe-oxyhydroxides and then released under reducing conditions in the Bay of Bengal is a model first developed by Bagla and Kaiser[8] and Bhattacharya *et al.*[10] and later adopted by other several Group.[5,67–69] Based on this hypothesis, As is scavenged and immobilized by Fe-oxyhydroxides in an aererated aquatic system, and release as a consequence of changing redox conditions. It can be released due to microbial activity that consum burried organic matter, breath Fe-oxyhydroxides and produced a dissolution of the Fe-oxyhydroxides under reduce conditions or related to the biodegradation of buried peat deposits causing reducing conditions in groundwater.

The fact that during the Holocene the climate is dominated by high rainfall conditions favouring the weathering of Himalayan rocks and have enhanced fluxes of finer Fe-hydroxides minerals, clay size detritus, particularly before 3000 yr BP. This can explained the enrichment of arsenic in Holocene aquifer sediments. Some authors proposed that As-bearing pyrite coming from the Gondwana coal seams and from the Rajmahal volcanics[5,11,12,67] while for Stanger,[39] the primary geogenic source of the arsenic has to be search in the Quamdo-Simao ophiolitic province in the central-eastern part of the Tibetan plateau. Finally Stüben *et al.*[7] proposed a diffuse sources of the origin of the As rather than a precise source. As the Bay of Bengal sediments are mainly feed by the Ganga-Brahmaputra river system since almost 20 Ma.[58] the origin of arsenic as to be search in the Himalayan system.

We propose that the Indus-Tsangpo suture zone dominated by arc-related rocks and more particularly by large volume of serpentinites enriched in arsenic could be the primary source of arsenic. The fact that the present day arsenic concentration in the main Himalayan river, and particularly the Siang-Brahmaputra river system is not so high as expected can be explained by strong aridic conditions present day prevailing in the Indus-Suture zone and do not favored the weathering of serpentinites into As rich-smectite and Fe-hydroxydes. Thus, we propose that the main arsenic transfer from the Himalayan system to the Bay of Bengal occurred during more humid periods.

The relationship between present-day arsenic anomalies in the river and in the groundwater of the Terai plain and the proximity of the mudy Siwalik thick deposits clearly suggest that the Siwalik Group is the main source of arsenic in the Ganga basin. Moreover, the important activity of the Main Frontal Thrust since the Holocene is probably an important factor explaining the enrichment of arsenic in the Ganga basin system. In fact, this thrust controlled the uplift and consequently the incision of the Siwalik Group leading to its preferential erosion. Combining rapid uplift of the Siwalik sediments with increasing runoff conditions during the Holocene are strongly favorable conditions to mobilize the arsenic initially scavenged in the fine-grained Siwalik clay levels and associated with iron mineralization, sulphides and clay minerals.

Although the Siwalik Group is probably the main arsenic reservoir in Himalaya, it cannot be the original source of this element. It is generally admit that the source of the detrital sediments feeding the Siwalik basin from 15 to 2 Ma is the Greater Himalaya.[28] However, the Himalayan barrier did not appear before 8 Ma[60] and the Himalayan hydrographic system strongly evolves till 5 to 3.5 Ma.[58,60] This suggests that the main Himalayan rivers before 5 Ma possibly cross all the Himalayan range from the Ladakh-Gangdese batholith, the Indus-Tsangpo suture zone to the Siwalik foreland basin (Fig. 5). Thus, arsenic-rich ophiolitic suture zone, exposed at the surface since a minimum of 40 Ma are probably strongly eroded during this period and we propose that they enriched the Siwalik foreland basin in arsenic.

Conclusion

The origin of arsenic enrichment in Holocene grounwater in the West Bengal and Bengladesh is highly debated. A review of the Himalayan system combined with its paleohydrology, tectonic and climatic evolution allow to propose that the original source of arsenic has to be searched in ophiolitic, arsenic rich, arc related rocks in the Indus-Tsangpo suture zone. The weathering of arc related rocks followed by the transport of arsenic into the Siwalik foreland basin, playing the role of arsenic reservoir from the Miocene to the Pleistocene. Intense tectonic activity in the front of the Himlayan belt associated with high rainfall conditions during the Holocene allow the arsenic to be remobilized, transport by the river and again immobilized and concentrated under oxidized conditions by Fe-oxyhydroxides and clay mineral in the Bay of Bengal.

The present remobilization of the arsenic is related to groundwater reduced conditions by microbial activity or interaction with biodegradation of buried organic matter. If this global scenario is correct, this suggests that the present-day normal concentration of arsenic measured in the Ganga-Brahmaputra-Meghna river system cannot explain the arsenic anomaly locally observed in the Bay of Bengal. Only an integrative global model taking into account the initial concentration of arsenic in potential source rocks, weathering processes, paleohydrology combining with tectonic and climate evolution and chemistry allow us to better understand the worst mass poisoning of a human population in history.

Acknowledgments

This work was supported by DYETI and EC2CO CNRS grants. We thank the three anonymous reviewers for their constructive comments.

References

[1] Acharyya, S.K.; Chackraborty, P.; Lahiri S.; Raymahashay, B.C.; Guha S.; Bhowmik, A. Arsenic poisoning in the Ganges delta. Nature **1999**, *401*, 545.

[2] Acharyya, S.K.; Lahiri, S.; Raymahashay, B.C.; Bhowmik, A. Arsenic toxicity of groundwater in parts of the Bengal basin in India and Bangladesh, the role of quternary stratigraphy and Holocene sea-level fluctuation. Environ. Geol. **2000**, *39*, 1127–1137.

[3] Charlet, L.; Polya, D. Arsenic in shallow reducing groundwaters in southern Asia, an environmental health disaster. Elements **2006**, *2*, 91–96.

[4] Das, D.; Samanta, G.; Mandal, B.K.; Chowdhury, T.R.; Chanda, C.R.; Chowdhury, P.P.; et al. Arsenic in groundwater in six districts of West Bengal, India. Envir. Geochem. Health **1996**, *18*, 5–15.

[5] Nickson, R.; McArthur, J.; Ravenscroft, P.; Burgess, W.; Ahmed, I.K.M. Mechanism of arsenic poisoning of groundwater in Bangladesh and West Bengal. Appl. Geochem. **2000**, *15*, 403–413.

[6] Nickson, R.; McArthur, J.; Shrestha, BR.; Kyaw-Myint, T.O.; Lowry, D. Arsenic and other drinking water quality issues, Muzaffargarh District, Pakistan. Appl. Geochem. **2005**, *20*, 55–68.

[7] Stüben, D.; Berner, Z.; Chandrasekharam, D.; Karamakar, J. Arsenic enrichment in groundwater of west Bengal, India: geochemical evidence for mobilization of As under reducing conditions. Appl. Geochem. **2003**, *18*, 1417–1434.

[8] Bagla, P.; Kaiser, J. Indias spreading health crisis draws global arsenic experts. Science **1996**, *274*, 174–175.

[9] Dixit, S.; Hering, J.G. Comparison of arsenic V and arsenic III sorption onto iron oxides minerals: implications for arsenic mobility: environmental science and technology. Environ. Sci. Tech. **2003**, *37*, 4182–4189.

[10] Bhattacharya, P.; Chatterjee, D.; Jacks, G. Occurrence of arsenic-contaminated groundwater in alluvial aquifers from Delta plain, Eastern India: options for a safe drinking water supply. Water Res. Dev. **1997**, *13*, 79–92.

[11] Ghosh, S.; De S. Sources of arseniferous sediments at Kauchua and Itina, Habra Block, North-24 Paragans, West Bengal, a case study. J. Earth Sci. **1995**, *22*, 183–189.

[12] Chandrasekharam, D.; Karamakar, J.; Berner, J.; Stüben, D. Arsenic contamination in groundwater, Murshidabad districy, West Bengal. In: Cidu R. (Ed). Proceeding 10th International Symposium on Water Rocks Interaction, Villasimius, Italy **2001**, *2*, 1051–1054.

[13] Korte, E.; Fernando, Q. A review of arsenic (III) in groundwater. Critical Rev. Environ. Control **1991**, *21*, 1–39.

[14] Milliman, J.D.; Sivitsli, J.P.M. Geomorphyc/tectonic control of sediment discharge to the ocean: the importance of small mountainous rivers. J. Geol. **1992**, *100*, 525–544.

[15] Garzanti, E.; Vezzoli, G.; Ando, S.; France-Lanord, C.; Singh, S.K.; Foster, G. Sand Petrology and focused erosion in collision orogens: the Brahmaputra case. Earth Planet. Sci. Lett. **2004**, *220*, 157–174.

[16] Summerfield, M.A.; Hulton, N.J. Natural controls of fluvial rates in major world drainage basins. J. Geophys. Res. **1994,** *99*, 13871–13883.

[17] Plant, J.A.; Kinniburgh, D.G.; Smedley, P.L.; Fordyce, F.M.; Klinck, B.A. Arsenic and Selenium. *Treatise on Geochemistry*, Lollar, B.S..; Holland, H.D.; Turekian, K.K., Eds **2003**, *9*, 17–66.

[18] Nath, B. Aquifers bearing low and High arsenic groundwater—a comparative study on litho and hydrogeochemistry in Chakdaha block, Nadia District, West Bengal. Ph-D thesis, University of Jadavpur, **2006**; 218p.

[19] BGS.; DPHE. Arsenic contamination of groundwater in Bangladesh. In Kinniburg, D.G.; Smedley, P.L. (Eds.), British Geological Survey Technical Report WC/00/19, British Geological Survey, Keyworth, **2001**.

[20] Stummeyer, J.; Marchig, V.; Knabe, W. The composition of suspended matter from Ganges-Bramaputra sediment dispersal system during low sediment transport season. Chem. Geol. **2001**, *185*, 125–147.

[21] Onishi, H.; Sandell, E.B. Geochemistry of arsenic. Geochim. Cosmochim. Acta **1995**, *7*, 1–33.

[22] Kansakar, D.R.; Geologic and geomorphologic characteristics of arsenic contaminated groundwater areas in Terai, Nepal. In: Kansakar, D.R. (ed) *Arsenic testing and finalization of groundwater legislation and finalization project*, summary project report, Latitpur, Nepal. H. M. Govt. of Nepal, Dept of Irrigation, **2004**; 31–47.

[23] Burbank, D.W.; Beck, RA.; Mulder, T. The Himalayan foreland basin. In Yin, A.; Harrison, T.M., Eds., *The Tectonic Evolution of Asia*: Cambridge Univ. Press **1996**; 149–188.

[24] Guillot, S.; Garzanti, E.; Baratoux, D.; Marquer, D.; Mahéo, G.; de Sigoyer, J. Reconstructing the total shortening history of the NW Himalaya. Geochem. Geophys. Geosyst., **2003**, *4*, XXXX, doi:10.1029/2002GC000484.

[25] Chakraborti, D.; Basu, G.K.; Biswas, B.K.; Choudhury, U.K.; Rahaman, M.M.; Paul, K.; et al. Characterization of arsenic bearing sediments in the gangetic delta of west Bengal, India. In: Chappe, W.R.; Abernathy, C.O.; Calderon, E.L., Eds., *Arsenic Exposure and Health Effects*, Elsevier **2001**; 27–52.

[26] Najman, Y.; Pringle, M.; Godin, L.; Oliver, G. The detrital record of orogenesis: a review of approaches and techniques used in the Himalayan sedimentary basins. Earth Sci. Rev. **2006**, *74*, 1–72.

[27] DeCelles, P.G.; Robinson, D.M.; Quade, J.; Ojha, T.P.; Garzione, C.N.; Copeland, P.; et al. Stratigraphy, structure, and tectonic evolution of the Himalayan fold-thrust belt in western Nepal. Tectonics **2001**, *20*, 487–509.

[28] Huyghe, P.; Mugnier, J.L.; Gajurel, A.P.; Delcaillau, B. Tectonic and climatic control of the changes in the sedimentary record of the Karnali River section (Siwalik of western Nepal). Island Arcs **2005**, *14*, 311–325.

[29] Gautam, N.; Fujiwara, Y. Magnetic polaritity stratigraphy of Siwalik Group sediments of Karnali river section in Western Nepal. Geophy. J. Int. **2000**, *142*, 812–824.

[30] Hisatomi, K. The sandstone petrography of the Churia (Siwalik) Group in the Arung Khola-Binai Khola rea, west central Nepal: Wakayama University of Natural Science. Bulletin Fac. Educ. **1990**, *39*, 5–29.

[31] Caillaud, J.; Proust, D.; Righi, D.; Martin, F. Fe-rich clays in a weathering profile developed from serpentinite. Clays Clay Min. **2004**, *52*, 779–791.

[32] Mugnier, J.L.; Huyghe, P.; Leturmy, P.; Jouanne, F. Episodicity and rates of thrust-sheet motion in the Himalayas (western Nepal). McClay, K.R., Ed., Thrust Tectonics and hydrocarbon system. AAPG Mem. **2004**, *82*, 91–114.

[33] Lavé, J.; Avouac, J.P. Active folding of fluviatile terraces across the siwalik hills, Himalayas of central Nepal, implications for Himalayan seismotectonics. J. Geophys. Res. 2000, *105*, 5735–5770.

[34] Chesley, J.T.; Quade, J.; Ruiz, J. The Os and Sr isotopic record of Himalayan paleorivers: Himalayan tectonics and influence on ocean chemistry. Earth Planet. Sci. Lett. **2000**, *179*, 115–124.

[35] Quade, J.; Roe, L.; DeCelles, P.G.; Ojha, TP. The late Neogne 87Sr/86Sr record of lowland Himalayan rivers. Science **1997**, *276*, 1828–1831.

[36] Shrestha, B.R.; Whitney, J.W.; Shrestha, K.B. The state of arsenic in Nepal. NASC/ENPHO **2003**; 122 p.

[37] Williams, V.S. Nepalese groundwater arsenic contamination is related to Siwalik source rock. Geol. Soc. Amer. Abs. Progr. **2005**, *37*, 170 p.

[38] Anonymous. The poison we drink. Business Standard (India) **2004**, 28th September 2004.

[39] Gurung, J.K.; Ishiga, H.; Khadka, M.S. Geological and geochemical examinaion of arsenic contamination in groundwater in the Holocene Terai Basin, Nepal. Environ. Geol., **2005**, *49*, 98–113.

[40] Stanger, G. A paleo-hydrogeological model for arsenic contamination in southern and south-east Asia. Environ. Geochem. Health **2005**, *27*, 359–367.

[41] Mahéo, G.; Bertrand, H.; Guillot, S.; Keller, F.; Capiez, P. The South Ladakh ophiolite (NW Himalaya, India), a crustal and upper mantle section of the same immature arc: implications for the closure of the Neothethys. Chem. Geol. **2004**, *203*, 273–303.

[42] Khan, A.M.; Jan, M.Q.; Weaver B.L. Evolution of the lower arc crust in Kohistan: temporal arc magmatism through early, mature and intra-arc rift stages. Himalayan Tectonics Treolar, P.J. and Searle, M. Eds., Geol. Soc. Sp. Pub. **1993**, *274*, 123–138.

[43] Quanru, G.; Guitang, P.; Zheng, L.; Chen, Z.; Fisher, RD.; Sun, Z.; et al. The Eastern Himalayan syntaxis: major tectonic domains, ophiolitic mélanges and geologic evolution. J. Asia. Earth Sci. **2005**, *27*, 265–285.

[44] Guillot, S.; Hattori, K.; de Sigoyer, J.; Nägler, T.; Auzende, A.L. Evidence of hydration of the mantle wedge and its role in the exhumation of eclogites. Earth Planet. Sci. Lett. **2001**, *193*, 115–127.

[45] Hattori, K.; Guillot, S. Volcanic fronts as a consequence of serpentinite dehydration in mantle wedge. Geology **2003**, *31*, 525–528.

[45] Le Fort, P.; Guillot, S.; Pêcher, A. HP metamorphic belt along the Indus suture zone of NW Himalaya: new discoveries and significance. C. R. Acad. Sci., Paris **1997**, *325*, 773–778.
[46] Hattori, K.; Takahashi, Y.; Guillot, S.; Johanson, B. Arsenate [As(V)] in serpentinites originated from the fore-arc mantle: X-ray absorption spectroscopy study. Geochim. Cosmochim. Acta **2005**, *69*, 5585–5596.
[47] Schlup, M.; Carter, A.; Cosca, M.; Steck, A. Exhumation history of eastern Ladakh revealed by 40Ar/39Ar and fission track ages: the Indus River-Tso Morari transect, NW Himalaya. J. Geol. Soc., London **2003**, *160*, 385–399.
[48] Pichler, T.; Veizer, J.; Hall, G.E.M. Natural input of arsenic into a coral-reef ecosystem by hydrothermal fluids and its removal by Fe(III) oxyhydroxides. Environ. Sci. Technol. **1999**, *33*, 1373–1378.
[49] Tapponnier, P.; Zhiqin, X.; Roger, F.; Meyer, B.; Arnaud, N.; Wittlinger, G.; et al. Oblique Stepwise Rise and Growth of the Tibet Plateau. Science **2001**, *294*, 1671–1677.
[50] White, N.; Pringle, M.; Garzanti, E.; Bickle, M.; Najman, Y.; Chapman, H.; et al. Constraints on the exhumation and erosion of the High Himalayan slab, NW India, from foreland basin deposits. Earth Planet. Sci. Lett. **2002**, *195*, 29–44.
[51] Yin, A. Cenozoic tectonic evolution of the Himalayan origen as constrained by along-strike variation of structural geometry, exhumation history, and foreland sedimentation. Earth Sci. Rev. **2006**, *76*, 1–131.
[52] Hodges, K.V.; Hames, W.E.; Olszewski, W.; Burchfiel, B.C.; Royden, L.H.; Chen, Z. Thermobarometric and 40Ar/39Ar geochronologic constraints on Eohimalayan metamorphism in the Dinggyê area, southern Tibet. Contrib. Mineral. Petrol. **1994**, *117*, 151–163.
[53] Vannay, J.C.; Hodges, K.V. Tectonometamorphic evolution of the Himalayan metamorphic core between the Annapurna and Dhaulagiri, central Nepal. J. Metam. Geol. **1996**, *14*, 635–656.
[54] DeCelles, P.G.; Gehrels, G.E.; Quade, J.; Ojha, T.P. Eocene-early Miocene foreland basin development and the history of Himalayan thrusting, western and central Nepal. Tectonics **1998**, *17*, 741–765.
[55] France-Lanord, C.; Derry, L.; Michard, A. Evolution of the Himalaya since Miocene time: isotopic and sedimentologic evidence from the Bengal fan. *Himalyan Tectonics*, Treolar, P.J. and Searle, M. Eds., Geol. Soc. London Spec. Pub. **1993**, *74*, 445–465.
[56] Alam, M.; Curray, J.R.; Chowdhury, L.L.R.; Gani, MR. An overview of the sedimentary geology of the Bengal Basin in relation to the regional tectonic. Sedim. Geol. **2003**, *155*, 227–270.
[57] Amano, K.; Taira, A. Two-phase uplift of Higher Himalayas since 17 Ma. Geology **1992**, *20*, 391–394.
[58] ugnier, J.L.; Huyghe, P. Ganges basin geometry records a pre-15 Ma isostatic rebound of Himalaya. Geology **2006**, *34*, 445–448.
[58] Brookfield, M.E. The evolution of the great river systems of southern Asia during the Cenozoic India-Asia collision: rivers draining southwards. Geomorphology **1998**, *22*, 285–312.
[59] Valdiya, K.S. Dynamic Himalaya. Universities Press, Hyderabad 1998, 178 p.
[60] Molnar, P.; England, P.; Martinod, J. Mantle dynamics uplift of the Tibetan plateau, and Indian monsoon. Rev. Geophys. **1993**, *3*, 357–396.
[61] Gartner, S.; Cochran, J.R.; Stow, D.A.. Neogene calcareous nannofossil biostratigraphy, Leg 116 (Central Indian Ocean). in Proceedings of the Ocean Drilling Programme, Cochran, J.R. and Stowe, D.A.V. Eds., O.D.P, College Station, TX, **1990**, 165–187.
[62] Clift, P. Controls on the erosion of Cenozoic Asia and the flux of clastic sediment to the ocean. Earth Planet. Sci. Lett. **2006**, *241*, 571–580.
[63] Mohindra, R. Holocene soil chronoassociation in part of the Middle Gangetic plain: morphological and micromorphological characteristics. Terra Nova **1995**, *7*, 305–314.
[64] Phadtare, N.R. Sharp decrease in summer monsoon strength 4000–3500 cal yr BP in the Central Higher Himalaya of India. Quatern. Res. **2000**, *53*, 122–129.
[65] Bhattacharya, A. Vegetation and Climate during the last 30,000 years in Ladakh. Palaeogeogr. Palaeoclimatol. Palaeoecol. **1989**, *73*, 25–38.
[66] Gasse, F.; Fontes, J.C.; Van Campo, E.; Wei, K. Holocene environmental changes in Bangong Co basin (Western Tibet). Palaeogeogr. Palaeoclimatol. Palaeoecol. **1996**, *120*, 792
[67] Nickson, R.; McArthur, J.; Burgess, W.; Ahmed, I.; K.M.; Ravenscroft, P.; Rahman, M. Arsenic poisoning of Bangladesh groundwater. Nature **1998**, *395*, 338.
[68] Appelo, C.A.J.; Van der Weiden, M.I.J.; Tournassat, C.; Charlet, L. Surface complexion of ferrous iron and carbonate of ferrihydrite and the mobilization of arsenic. Environ. Sci. Technol. **2002**, *36*, 3096–3103.
[69] Ravenscroft, P.; McArthur, J.M.; Hoque, B.A. Geochemical and palaeohydrological controls on pollution of groundwater by arsenic. In: Arsenic Exposure and Health Effects IV, Chappell, W.R.; Abernathy, C.O.; Calderon, R.L., Eds, Elsevier, Oxford. **2001**; 53–77.