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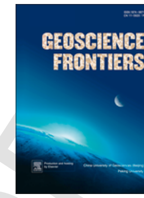

Research paper

Geochemistry of Harappan potteries from Kalibangan and sediments in the Ghaggar River: Clues for a dying river

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ABSTRACT

The ephemeral Ghaggar-Hakra River of north-western India has always been considered to be the remnant of an ancient perennial glacier-fed river (Vedic Saraswati). The exact reason and timing of major hydrological change of this river remains speculative. The river's purported association with the zenith of the Harappan civilisation remains a conjecture because the timings of its fluvial past are still being debated. In this study we have made an attempt to resolve this issue using geochemical provenance of sediments from some dated horizons in the Ghaggar flood plain and that of the material used in the potteries from the Mature Harappan period (4600 to 3900 yr BP) at Kalibangan. Sampled sedimentary horizons were dated by radiocarbon and optically stimulated luminescence (OSL) methods. Results of our study from the Ghaggar alluvium indicate that the river did have glacial sources during the early Holocene. However, the data from the potteries suggest that during the Mature Harappan period, the sediments in the Ghaggar as used by the potters did not have a higher Himalayan provenance and hence, were not derived from glaciated Himalayas. These findings imply that during the time of the Mature Harappans the Ghaggar had already become a foothill-fed river.

1. Introduction

Rivers have always played major roles in shaping the history of mankind. They are the silent witnesses of rise and fall of civilisations across the globe, and have remained part of the cultural identity of these civilisations. One such fascinating example has been the story of a supposedly mighty river in the heart of the Bronze Age Harappan settlements and its purported link to the evolution of the civilisation. More than a century of research have confirmed existence of several paleo-channels along the present day, mostly defunct Ghaggar-Hakra streams in the semi-arid northern margin of the Thar Desert (Fig. 1A and B; Valdiya, 2013). However, no conclusive evidence has yet been provided establishing a link between the paleo channels and a pre-historic, glacial fed river system. Many hypothesise that these paleo-channels are the remnants of the mythical river Saraswati, which was first described in the three millennia old scriptures of Rig-Veda (Oldham, 1893; Ghose et al., 1979; Pal et al., 1980; Radhakrishan and Merh, 1999; Kochar, 2000; Valdiya, 2013). It is well known that a great majority of Harappan settlements are concentrated along the present-day dry beds of the Ghaggar-Hakra channels (Mughal, 1997; Possehl, 2002; Fig. 1A). Since availability of water is one of the key requirements for development of civilisations, it is quite natural to believe that the Ghaggar-Hakra streams had a strong fluvial past during the peak of the Indus Valley civilisation. Hypotheses based on this idea suggest that the drying up of the Ghaggar-Hakra River, owing to river reorganisation, triggered the abrupt decline of the Indus Valley tradition in the north-western India, four millennia ago (Misra, 1984; Mughal, 1997; Possehl, 2002; Kenoyer, 2008; Wright et al., 2008). However, lack of geochronological data makes it difficult to constrain the exact timing of these events. Competing hypotheses, based on geochemical studies, propose that the Ghaggar-Hakra system had already become rain-fed (by ~10 ka) by the time the early Harappans had started settling down in its flood plain (Clift et al., 2012; Giosan et al., 2012; Tripathi et al., 2013), and that subsequent climate change leading to extreme aridity sealed the fate of the civilisation (e.g. Sarkar et al., 2016). Therefore, the issue of a possible glacial past of the Ghaggar-Hakra river system remains unresolved. Even if the river did have a glacial history, the timings and reasons of the hydrological changes remain to be unravelled.

Apart from indecipherable information about their lifestyle, the Indus valley people had left behind a plethora of artefacts including sophisticated potteries and ceramics. These have the potential to reveal a lot about their living environments. It has been observed that ancient potters generally used materials available in their near geographical vicinity to create potteries (Krishnan, 2002). In the case of the Indus valley potteries, source material would have been the abundant flood-plain sediments deposited by the rivers, on which the cities were built. Thus, geochemical composition of potteries is likely to give us an insight into the sediment composition of the river during that period. This in turn can reveal about the catchment of the river,

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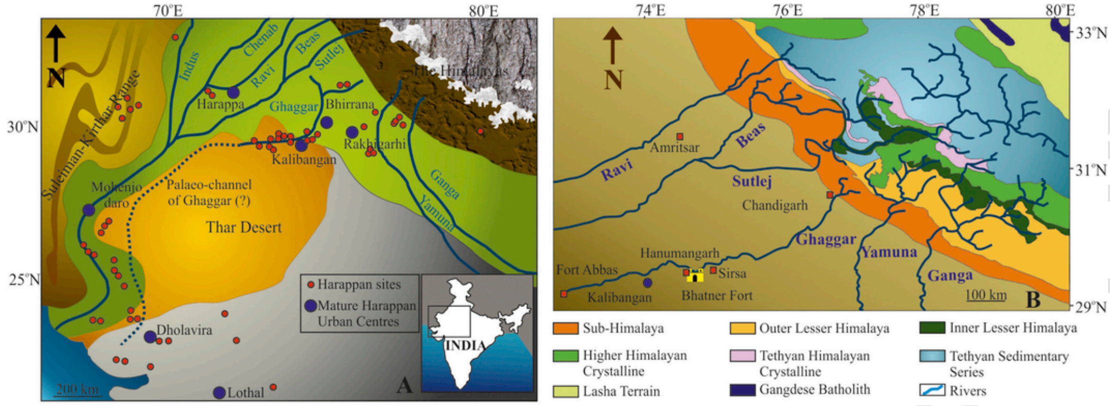

Figure 1. (A) Regional geomorphological map of north-western India showing the major landscapes. The putative course of paleo-Ghaggar is shown as dotted line and the pre-historic Harappan settlements are shown. Map is modified after Sarkar et al. (2016). (B) Different litho-tectonic units of the Himalayas from where different western Indian rivers have originated. Also shown are the positions of Kalibangan and Bhatner Fort on the bank of the Ghaggar. Map is modified after Singh et al. (2016).

glaciated higher Himalayas versus piedmont origin, because the sediment in the Indo-Gangetic plains are geochemically well co-related with their sources in the Himalayas (Najman, 2006). However, this method of source fingerprinting has its own limitations because of overlapping chemical compositions of the sources, differential chemical weathering at the source and in the alluvial plain, and alteration of original signals due to recycling. In spite of these, isotopic fingerprinting has remained the most promising tool for provenance study in the sediments derived from the Himalaya (Najman, 2006). In the present study, for the first time, we have made an attempt to address the issue of the lost river Saraswati from an entirely new angle, using geochemistry of well-dated Ghaggar alluvial deposits and comparing them with that of the potteries of the Mature Harappan period (4600–3900 yr BP), recovered from the Harappan acropolis of Kalibangan.

2. Harappan settlements along Ghaggar

The Indus Valley/Harappan cultural tradition developed along the river valleys of the Indus and Ghaggar-Hakra during mid-Holocene (Fig. 1). People of this culture settled over an area larger than the contemporaneous Mesopotamian and Egyptian civilisations (~8,00,000 km^2; Danino, 2010). The duration of existence of this culture, based on dates from Harappa and nearby localities, have been divided into four phases/periods (Kenoyer, 1998; Wright et al., 2008; Dikshit, 2013). Around 5.7 kyr BP agro-pastoral Ravi culture flourished, followed by the transitional Kot Diji Phase (~4.8 kyr BP). The sophisticated urban civilization of the Mature Harappan phase started around 4.6 kyr BP and disintegrated at ~3.9 kyr BP, followed by a de-urbanisation era of Late Harappan phase that lasted until ~3.3 kyr BP. However, Possehl (2002) had proposed a much older age for the Harappan culture based on spatio-temporal distribution of archaeological remains that spread across the Indian sub-continent. Earlier, Mughal (1997) had reported such older pre-Harappan settlements along the Hakra river of Cholistan desert and named it as the Hakra Phase. Later, numerous sites of the Hakra phase were discovered along the dry beds of the Ghaggar-Hakra including Kalibangan (the present study site), Farmana, Bhirrana, Rakhigarhi etc. Based on available chronological information the antiquity of the Hakra Phase can be pushed back to ~9.5 kyr BP (Sarkar et al., 2016 and references therein).

Kalibangan is one of the important Harappan cities situated on the southern bank of the river Ghaggar (Fig. 3) and has a continuous history since the Hakra Phase, up to the Late Harappan (Thapar, 1975). The oldest dated sequence of Kalibangan is 7.6 kyr BP (Sarkar et al., 2016). The settlement has two fortified sections, the Citadel (KLB-I) and the lower city (KLB-II) located to the east of the citadel (Fig. 2A). The KLB-I mound contains ruins of the settlements since the Hakra Phase until the late Harappan phase, whereas the KLB-II mound is characterised by only the mature Harappan settlements. Fig. 2B shows the mound of KLB-II as photographed during our field work in 2014. The remains of brick walls and terracotta pipelines can be seen in Fig. 2C. To understand the source of the clay used in making the potteries and bricks by the Mature Harappans we restricted our sampling to the KLB-II mound. Photographs of some of these samples are shown in Fig. 2D.

Giosan et al. (2012) had suggested that subsequent to de-urbanisation, Harappan settlements gradually shifted north-eastward to the upper Haryana plains. The Ghaggar-Hakra valley was later re-occupied by Painted Grey Ware sites during 3000–2600 kyr BP. Also during the medieval period fortifications were made along these floodplains (Mughal, 1997). The Bhatner Fort (12th century AD) of Hanumangarh is one of them (Fig. 1B). For the present study we also sampled bricks from this fort.

3. Methodology

Optically Simulated Luminescence (OSL) dating of the sand horizons was done in Physical research Laboratory using single aliquot regeneration (SAR) protocol of Murray and Wintle (2000). One sample of gastropod shells collected from a silty-clay horizon was dated by AMS C-14 at Centro Nacional de Aceleradores (CNA), Spain. The C-14 date have been calibrated using INTCAL 13 curve of Reimer et al. (2013).

All the geochemical and isotopic measurements were done on carbonate free siliciclastic/silicate fractions of samples. Samples were washed with 18.2 MΩ cm water multiple times, dried at 110 °C, powdered and homogenised. Prior to dissolution, powdered samples were heated at 650 °C for 2 h to remove organic matter and leached in 2 N HCl to remove carbonates. ~50 mg each of de-carbonated powdered samples was dissolved using standard HF-HNO_3 (2:1) acid digestion technique. 15 mL Savillex Teflon vials were used for the purpose. Complete dissolution was achieved through repeated ultrasonication, heating, aqua regia and 8 N HNO_3 treatments. It was made sure that no residue was left. Final (stock) solutions were prepared in 0.2 N HNO_3. Ultra pure (low blank) acids were used for the purpose.

Concentrations of trace elements including rare earth elements (REEs) were measured using a Thermo X-Series 2 Q-ICPMS at

Figure 2. (A) The settlement map of the Harappan acropolis of Kalibangan. Map is modified after Thapar (1975). (B) The KLB-II mound of Kalibangan as photographed during field work in 2014. (C) The remains of brick walls can be seen through the gaps in the mound. In the inset image terracotta drainage pipes can be seen. (D) Samples of Mature Harappan potteries collected from the KLB-II mound.

Physical Research Laboratory (PRL). USGS rock standard BHVO-2 was used as unknown for accuracy check. Reproducibility of trace element contents, based on repeated analyses of the standard, was ≤3% for REEs and ≤6% for all other trace elements at 2σ level. Sr separation was done by conventional cation exchange column chemistry using AG®50W-X8 resin and Nd was separated from other REEs using Ln-specific resin from Eichrom® with dilute HCl as elutant (Dickin, 2000; Awasthi et al., 2014). $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ were measured in static multicollection mode on an IsoProbe-T TIMS (Ray et al., 2013). The measured isotopic ratios were corrected for fractionation using $^{86}Sr/^{88}Sr = 0.1194$ and $^{146}Nd/^{144}Nd = 0.7219$, respectively. The average values for NBS987 and JNdi measured over a period of 5 years are $^{87}Sr/^{86}Sr = 0.71023 \pm 0.00001$ ($n = 70$) and $^{143}Nd/^{144}Nd = 0.512104 \pm 0.000004$ ($n = 60$; ±0.1 in ε_{Nd} units) at 2σ level of uncertainty. The value of $^{143}Nd/^{144}Nd = 0.512104$ for JNdi corresponds to a value of 0.511847 for the widely used La Jolla Nd standard (Tanaka et al., 2000). $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ for BHVO-2 were 0.70346 ± 0.00004 and 0.512967 ± 0.000008 ($n = 10$; ±0.2 in ε_{Nd} units at 2σ), respectively, which are same as the reported values of 0.70344 ± 0.00003 and 0.51296 ± 0.00004 within 2σ (Raczek et al., 2001). To compare our data with that from literature, all $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios were normalized to 0.71025 for NBS987 and 0.511858 for La Jolla, respectively. All plots and discussion below are based on the normalized ratios.

4. Results and discussion

4.1. Sedimentary facies and depositional ages

The present course of the Ghaggar River is almost non-existent beyond Sirsa in Haryana due to damming and heavy farming on and along its shallow channel. The flood plain topography is monotonously flat and is bordered by sand dunes. Interestingly, the subsurface sedimentary facies is very different from that of the dry river bed. Fig. 3 presents a comparison of the subsurface stratigraphy from different localities along the flood plain as determined by us (Fig. 3A and B) and Saini et al. (2009) (Fig. 3C and D). The modern surface mud covers the Ghaggar alluvium near the present course of the Ghaggar channel. The older floodplain deposit of brown coloured silty-clay directly underlies the surface mud. The brown silty-clay directly overlies either a yellowish-brown fine fluvial sand layer or a grey micaceous sand layer.

The thickness of the overlying brown silty-clay varies from 1 to 10 m. AMS C-14 dating of gastropod shells from this horizon near Hanumangarh yielded an age of 3109 ± 35 cal yr BP, which can be considered as the age of deposition of the layer (Fig. 3A). Earlier Saini et al. (2009) had reported OSL depositional ages of 2.9 ± 0.2 kyr BP and 3.4 ± 0.2 kyr BP for the similar stratigraphic horizons around Sirsa (Fig. 3C and D). The silty-clay layers can be traced further downstream until the river bed vanishes in the desert. In deeper sections the silty-clay facies can be observed as intermittent thin layers stacked within the thick sand horizons deposited around 7–5 kyr BP (Clift et al., 2012; Singh et al., 2016). Based on the available chronological information it can be suggested that even though the deposition of the silty-clay sediments started around mid-Holocene, it had become a dominant facies by 3 kyr BP.

A detailed clay mineralogical study has been conducted by Alizai et al. (2012) which characterises the clay depositions of the Ghaggar alluvium, further downstream at Fort Abbas, Marot and Tilwala in Pakistan (Fig. 4). Considering that no tributaries join the Ghaggar downstream beyond Shatrana, it can be inferred that the clay mineralogical composition should have remained similar all along the floodplain. As suggested by Alizai et al. (2012) the most abundant clay mineral in the Ghaggar alluvium is smectite (51–59%), followed by illite (30–37%). The minor constituents are chlorite (5–7%) and kaolinite (2–5%). It can be observed in Fig. 4 that the abundances of these four clay minerals had remained spatially and temporally invariant during the Holocene. The presence of illite as a major clay indicates that the sediments were sourced from the Himalaya where physical weathering dominates. On the other hand dominance of smectite, which is primarily a product of chemical weathering, is not in accordance with a Himalayan source where chemical weathering is very less. Such a scenario can be explained by two-cycle weathering (Singh et al., 2005), one at the source and the other within the floodplains with smectite being generated in the latter as a result of chemical weathering.

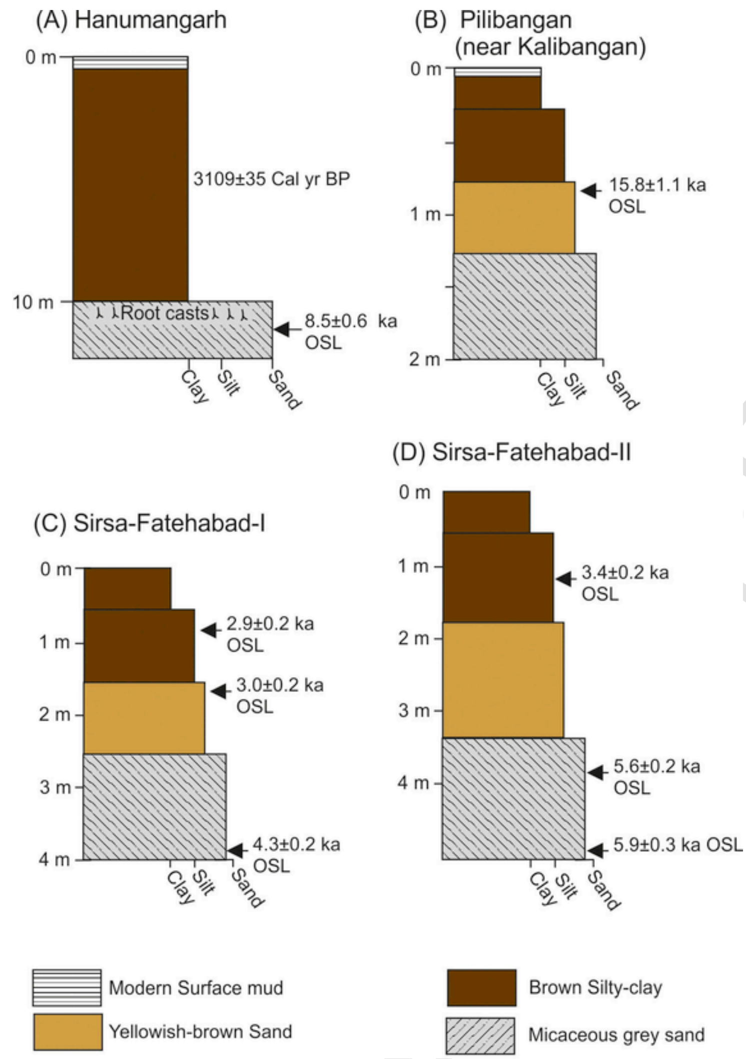

Figure 3. The subsurface facies architecture of the Ghaggar alluvium at (A) Hanumangarh, (B) Pilibangan and (C) Sirsa-Fatehbad-I and (D) Sirsa-Fatehbad-II regions. The stratigraphies of Sirsa-Fatehbad region are from Saini et al. (2009).

Raikes (1968) first reported coarse grained micaceous grey sand facies, resembling the sediment carried by modern higher Himalayan glacier-fed rivers like Ganga and Yamuna, buried ~10 m below the silty-clay floodplain deposits of the Ghaggar near Kalibangan. During the field survey for the present study similar grey sand was encountered at several places along the Ghaggar floodplain. Several other workers have also reported such sub-surface sand horizons from many locations in the floodplain (Saini et al., 2009; Saini and Mujtaba, 2012; Singh et al., 2016). The immature character of these coarse grained sand layers bears the testimony of a once active major fluvial system and they probably represent bedload sediments. Presence of a sharp contact between the micaceous sand and silty-clay layers points to a depositional hiatus/erosional surface between the two. Quartz is the most dominant mineral in the grey sandy facies, followed by feldspar and muscovite. Accessory phases include biotite, amphibole, kyanite, sillimanite, garnet and pyroxene (Saini et al., 2009). The clay content of the grey micaceous sand is almost negligible implying that these were high energy depositions. OSL dating of this sand layer at Hanumangarh yielded a depositional age of 8.5 ± 0.6 ka (Fig. 3A). The youngest age for similar grey sand from the upstream part of the river is reported to be 4.3 ± 0.2 ka (Saini et al., 2009; Fig. 3C). Singh et al. (2016) had reported depositional timing of these grey micaceous sands from Kalibangan region to be from ~70 to ~20 kyr BP.

At other locations, the grey sand is overlain by yellowish-brown fluvial sands which gradually grades upward into the brown silty-clay. This fine fluvial sand appears to have been deposited by a weaker phase of fluvial activity and local sediment reworking from dunes (Saini et al., 2009; Saini and Mujtaba, 2012). The OSL age of this yellowish-brown fluvial sand from the Pilibangan region (near Kalibangan), determined by us, is 15.8 ± 1.1 ka (Fig. 3B), whereas, Saini et al. (2009) had reported much younger depositional age of 3 ± 0.2 ka for similar deposits near Sirsa (Fig. 3C). These young sand layers (younger than the grey sand) were likely deposited during a weaker phase of the river when suspended load dominated the system. Mineralogically, these sand deposits are predominantly composed of quartz and feldspar. Unlike the grey sandy facies, mica is less abundant and occurs as fine round-edged grains (recycled).

A graphical representation of temporal dominance, during the last 70 kyrs, of different sedimentary facies within the Ghaggar flood plain is given in Fig. 5. The age data from earlier studies and those presented here suggest that during early Holocene both types of sands

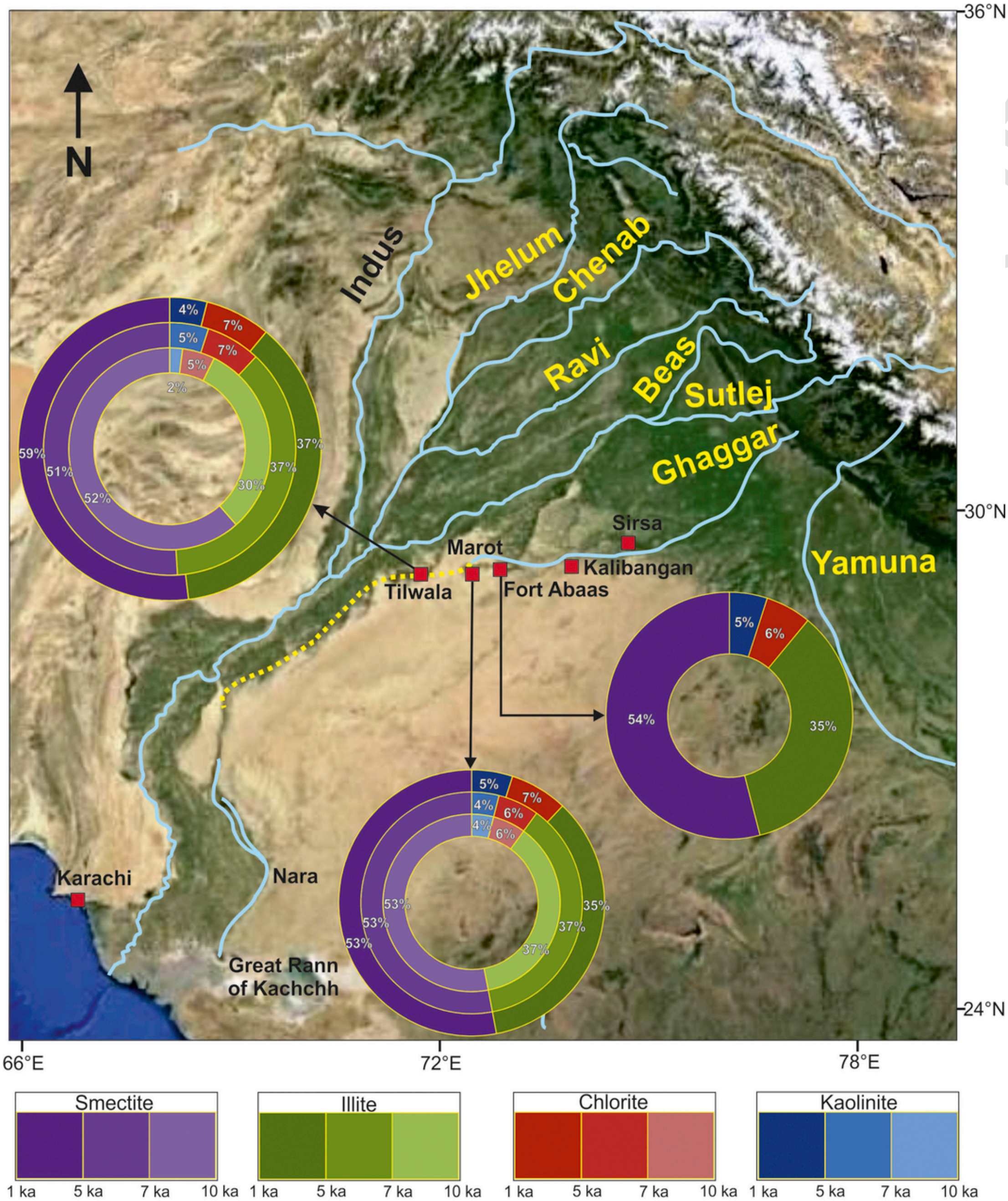

Figure 4. Pie chart showing the compositional variations of clay minerals across the Ghaggar alluvium during the Holocene.

(coarse grey and fine yellowish-brown) were getting deposited in the Ghaggar valley, whereas the sedimentation was dominated by only the grey sand during earlier times (Singh et al., 2016). This implies that the Ghaggar River was a much stronger fluvial system in the past and gradually, over a period of ~10 kyrs, reduced into a dwindling meandering system dominated with reworked and suspended sediments. For proper characterization of the sources of the sediments and to understand the depositional pathways, we studied the geochemical properties of these sediments along the Ghaggar flood plain near Hanumangarh and Pilibangan.

4.2. Geochemistry of Ghaggar alluvium

The trace element data of sediment samples are presented in Table 1 and plotted in Post Archean Australian Shale (PAAS) normalized diagram in Fig. 6. As can be seen, all different types of sediments show similar trace element patterns. Even the modern surface mud deposited during the latest flooding event shows a similar pattern. The only difference between these is the elemental concentrations. The modern mud in the river has the highest trace element contents, whereas the oldest alluvium, the coarse grey sand, has the lowest. This can be attributed to effect of dilution because of presence of

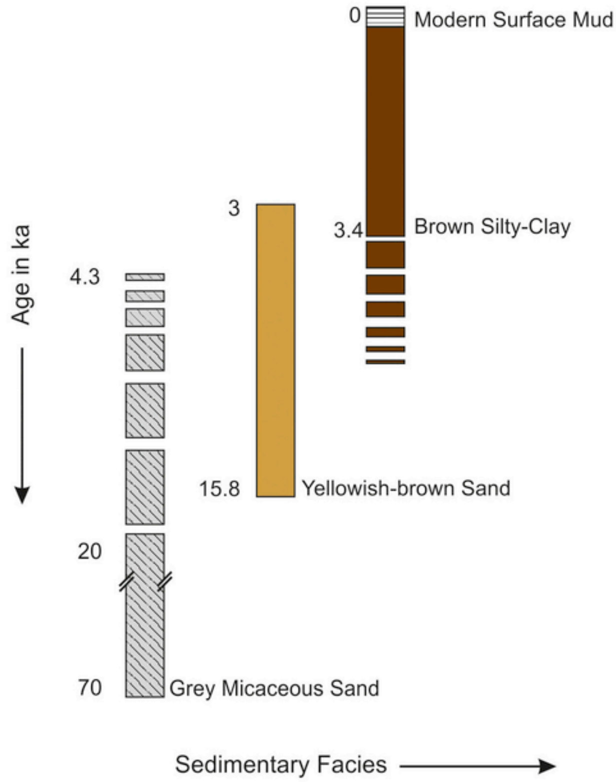

Figure 5. A graphical representation of composite stratigraphy of different sedimentary facies deposited within the Ghaggar floodplain over the past 70 kyr.

abundant quartz in the latter. Notwithstanding the differences in the contents, comparable patterns of trace elements in different sediments point to their derivation from analogous sources. The strong negative PAAS-normalized anomalies observed for Zr and Hf in the Ghaggar alluvium (Fig. 6) has also been reported from the sediments in the five rivers of Punjab (Alizai et al., 2011). Such a resemblance points to a similarity of provenances of sediments in Ghaggar and the rivers of Punjab, i.e., the Himalayas. The above depletions in the Zr and Hf could also be attributed to the removal of heavy mineral such as zircon from the sediments during long distance transport (Carpentier et al., 2009; Chatterjee and Ray, 2017), however, we do not envisage such a scenario considering that the sampled locations in all the rivers were not very far from the catchment. Therefore, the only likely possibility remains is that the sources of these sediments already had these depletions. Considering that the sources were located in the Himalayas, it needs to be determined if they were located either in the Higher Himalaya, or the Lesser Himalaya, or the Siwaliks.

To resolve the above issue we took the help of Sr—Nd isotopic composition of bulk sediments. The isotopic data of the Ghaggar alluvium are presented in Table 2. Different litho-tectonic units of the Himalayas are well characterised with respect to Sr and Nd isotopic compositions and can be used for tracing the provenance of the sediments in frontal alluvial plain. The Sr—Nd isotopic ratios of different Himalayan litho-tectonic units, based on the available data from literature, are shown in Fig. 7A. The glaciated regions of the Himalayas comprise rocks of the Higher Himalayan Crystalline Series (HHCS) and Lesser Himalayan Series (LHS). Rivers originating from the glaciers carry sediments derived from these two sources and hence they possess a mixed signal. In Fig. 7B we compare the Sr—Nd isotopic data for the Ghaggar alluvium, ours as well as that from literature (Tripathi et al., 2013; Singh et al., 2016), with that of the sub-Himalayan lithologies (Subathu, Kasauli, Dagsahi and Siwaliks) and for sediments in the rivers originating from the higher Himalaya

Table 1
Trace element data (in ppm) for samples from the Ghaggar flood-plain.

Sample	HG-14-4 (Brown silty-clay)	HG-14-8 (Brown silty-clay)	HG-14-16 (Brown silty-clay)	HG-14-18 (Modern surface mud)	HG-14-20 (Brown silty-clay)	HG-14-21 (Brown silty-clay)	HG-14-22 (Yellowish-brown sand)	HG-14-39 (Brown silty-clay)	HG-14-17 (Grey sand)	BHVO-2 (Measured)	BHVO-2 (Reported)
Cs	7.49	8.98	6.67	9.14	9.67	6.48	3.38	7.78	3.40	0.12	0.1 ± 0.01
Rb	124.1	145.2	113.7	151.9	148.7	109.7	76.0	115.6	74.2	10.3	9.11 ± 0.04
Ba	501	565	414	546	589	493	396	396	292	130	131 ± 1
Th	20.0	15.0	13.7	20.7	15.9	14.0	10.4	13.6	15.1	1.1	1.22 ± 0.06
U	2.27	1.34	1.54	2.50	2.91	2.02	1.37	1.69	1.57	0.42	0.403 ± 0.001
Nb	14	13	12	14	13	11	9	14	8	17	18.1 ± 1
Ta	1.11	1.04	0.93	1.14	1.16	0.86	0.71	1.03	0.75	0.95	1.14 ± 0.06
La	51	42	36	49	38	36	33	40	37	15	15.2 ± 0.1
Ce	99	86	73	98	76	72	67	79	73	39	37.5 ± 0.2
Pb	22.1	25.0	21.2	25.7	21.3	18.7	18.5	13.8	15.4	1.2	1.6 ± 0.3
Pr	11.9	9.8	8.3	11.2	8.6	8.3	7.6	9.0	8.2	5.2	5.35 ± 0.17
Sr	72	72	69	98	147	159	179	109	69	388	396 ± 1
Nd	43	36	31	41	31	30	28	32	29	24	24.5 ± 0.1
Zr	17	19	9	21	17	14	11	20	10	163	172 ± 11
Hf	0.6	0.7	0.6	0.8	0.8	0.5	0.4	0.7	0.3	3.9	4.36 ± 0.14
Sm	8.1	6.7	5.7	7.6	5.5	5.5	5.0	5.7	5.3	5.9	6.07 ± 0.01
Eu	1.4	1.3	1.1	1.4	1.0	1.0	1.0	1.0	0.8	2.0	2.07 ± 0.02
Gd	7.0	5.7	4.9	6.7	4.7	4.6	4.3	4.8	4.5	6.1	6.24 ± 0.03
Tb	0.81	0.69	0.60	0.82	0.56	0.55	0.52	0.57	0.53	0.84	0.92 ± 0.03
Dy	4.1	3.6	3.2	4.1	2.9	2.9	2.7	2.9	2.7	5.2	5.31 ± 0.02
Y	19.4	16.9	13.7	18.8	10.7	14.5	13.9	14.1	13.4	23.4	26 ± 2
Ho	0.70	0.64	0.55	0.72	0.51	0.51	0.50	0.50	0.46	0.89	0.98 ± 0.04
Er	1.9	1.8	1.6	2.0	1.5	1.4	1.4	1.4	1.3	2.5	2.54 ± 0.01
Tm	0.2	0.2	0.2	0.3	0.2	0.2	0.2	0.2	0.2	0.3	0.33 ± 0.01
Yb	1.6	1.5	1.3	1.6	1.3	1.2	1.2	1.2	1.1	1.9	2 ± 0.01
Lu	0.20	0.20	0.17	0.22	0.18	0.17	0.17	0.16	0.14	0.26	0.274 ± 0.005

Reproducibility (2σ): REE ≤ 3%; others ≤ 6%. Reported values for BHVO-2 are from Jochum et al. (2005).

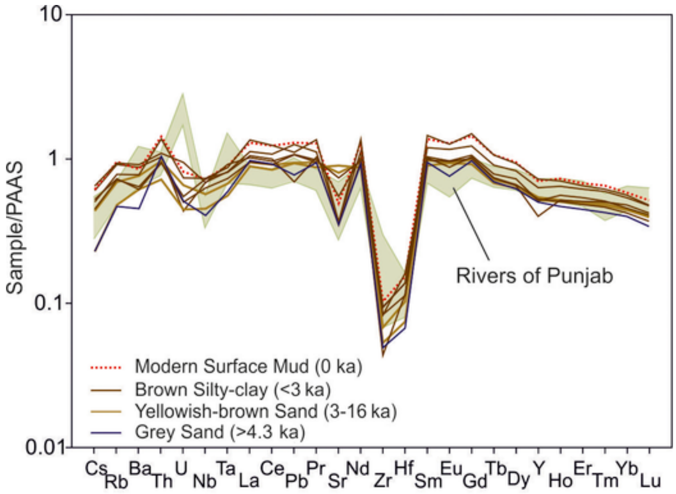

Figure 6. PAAS normalised trace element distribution of various sedimentary facies from the Ghaggar alluvium. The green coloured field in the background shows the range of composition observed in the rivers of Punjab. Data source: Alizai et al. (2011).

(Ganga, Yamuna and Sutlej; Singh et al., 2008; Tripathi et al., 2013). The micaceous grey sand layers near Kalibangan (>20 kyr BP; Singh et al., 2016) have overlapping isotopic compositions with that of the sediments in most glacier fed rivers, thus suggesting a provenance in the glaciated Himalayas (Fig. 7B).

The brown coloured silty-clay and the yellowish-brown fluvial sand possess distinctly different isotopic ratios than that of the grey sand (Fig. 7B); implying that the provenance of the brown silty-clay and yellowish brown sand is different from that of the Higher-Himalaya originated grey sands. The brown silty-clay and the yellowish-brown sand are less radiogenic in Sr and more radiogenic in Nd isotopic composition. They are also different in composition from the surrounding sand dunes, indicating very little, if any, input from the dunes via reworking. Fig. 1B shows that the modern river Ghaggar has its catchment in the sub-Himalayas which includes the Siwalik Group, Kasauli, Dagsahi and Subathu Formations. Consequently, the river is expected to carry sediments derived from these lithologies. Sr—Nd isotopic compositions of these lithologies are shown in Fig. 7B. Tripathi et al. (2013) have argued for a significant contribution of the Subathu Formation in the Ghaggar Alluvium. However, our observations suggest that the Subathu Formation having very different isotopic compositions might have had very little influence on the Ghaggar sediments (Fig. 7B). It appears that the rocks of the Siwalik Group, Kasauli and Dagsahi Formations are the major sources for the brown mud and yellowish-brown sand of the Ghaggar flood-plain (Fig. 7B). The more radiogenic Nd of the marginal desert dunes can very well be the results of sediment mixing from the river Indus. Given the fact that the sedimentation in the Ghaggar was dominated

Table 2
Isotopic data for sediment samples and archaeological artefacts from the Ghaggar flood-plain.

Samples	Brown Silty-clay (<3 ka)				Yellowish brown sand		Modern surface mud	
	HG-14-4	HG-14-8	HG-14-16	HG-14-20	HG-14-21	HG-14-22	HG-14-18	HG-14-18R
$^{87}Sr/^{86}Sr$	0.743022	0.745287	0.747306	0.733185	0.731733	0.730670	0.738182	0.738182
ε_{Nd}	−14.7	−14.6	−14.8	−14.3	−14.3	−13.4	−14.1	−13.8

Samples	Thar Dune sand (sub-surface, no age control)					
	HG-14-23	HG-14-24	RM-14-1	RM-14-3	HG-15-7	HG-15-32
$^{87}Sr/^{86}Sr$	0.726126	0.726132	0.727257	0.727815	0.729716	0.727714
ε_{Nd}	−13.0	−10.3	−10.9	−13.3	−13.8	−13.4

Samples	Kalibangan Pottery and brick (4.6–3.9 ka)				Bhatner Fort Brick (~0.9 ka)
	KBP-1	KBP-2	KBP-3	KBP-4	HGP-1
$^{87}Sr/^{86}Sr$	0.739043	0.731819	0.737543	0.726857	0.730976
ε_{Nd}	−12.4	−14.2	−13.1	−13.5	−12.9

$\varepsilon_{Nd} = [(^{143}Nd/^{144}Nd)_{sample}/(^{143}Nd/^{144}Nd)_{Chondrite}-1] \times 10^4$. Reproducibility ($2\sigma$): $^{87}Sr/^{86}Sr = \pm 0.000005$; $\varepsilon_{Nd} = \pm 0.1$.

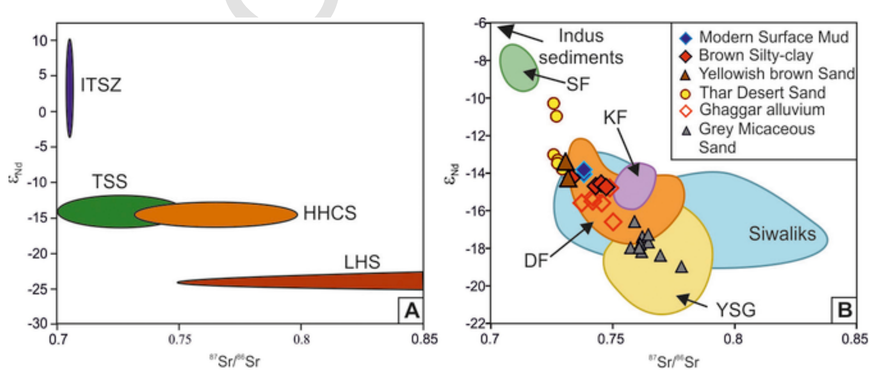

Figure 7. (A) ε_{Nd} vs. $^{87}Sr/^{86}Sr$ plot of major Himalayan litho-tectonic units showing their range of values. Data: Najman et al. (2000). ITSZ: Indo Tsangpo Suture Zone; TSS: Tibetian Sedimentary Series; HHCS: Higher Himalayan Crystalline Series; LHS: Lesser Himalayan Series. (B) ε_{Nd} vs. $^{87}Sr/^{86}Sr$ plot of various types of sediments from the Ghaggar alluvium compared with the sub-Himalayan provenances. Data for Ghaggar alluvium (red open diamonds) and grey micaceous sand (grey triangles) are from Tripathi et al. (2013) and Singh et al. (2016) respectively. SF: Subathu Formation; KF: Kasauli Formation; DF: Dagsahi Formation; YSG: Yamuna-Sutlej-Ganga sediment.

with suspended material by ~3 kyr BP, it can be suggested that the river likely had become a foothill fed river similar to its present condition by then.

From our geochemical provenance study it is clear that the Ghaggar had changed from a glacier fed strong fluvial system to a rain fed alluvial river during the Holocene. Interestingly, there is no geomorphic evidence of a glacial origin of the river. Therefore, the only plausible pathway for the higher Himalayan sediments to reach the Ghaggar channel would have been through the neighbouring rivers; the Sutlej and Yamuna, which used to flow into the Ghaggar during the pre-historic times (Valdiya, 2017 and references therein). This might explain the strong fluvial past of the river. The reason for the gradual hydrological changes in the Ghaggar could have been due to the progressive migration of these glacial-fed tributaries (Sutlej or Yamuna) away from it.

4.3. Isotopic fingerprinting of Kalibangan potteries

Potters of Bhirrana, a Harappan acropolis on the bank of the Ghaggar, used to make earthenware using clay from nearby localities (Krishnan et al., 2012). Extending this finding to Kalibangan it could be argued that potters here too had utilized the silty-clay which was available aplenty in the nearby Ghaggar floodplain. The very fact that common clay (illite/smectite, kaolinite and micas) can be utilized for general ceramics (Valášková, 2015) it is highly likely that the Harappans at Kalibangan made use of locally available clays, the mineralogical details of which are shown in Fig. 4 and discussed in Section 4.1. The usability of these silty-clay horizons is very much evident even today in the numerous active brick kilns all along the Ghaggar floodplain.

Another important understanding of ancient pottery making is that pure clay was never used for the purpose (Krishnan and Rao, 1994; Krishnan, 2002). For strengthening and creating different textures, various amounts of coarser material, generally sand, were mixed with pure clay to prepare the raw material. Therefore, one expects to find mixed geochemical signatures of sand and clay of the Ghaggar flood plain in the Kalibangan potteries. Fig. 8 presents ε_{Nd} versus $^{87}Sr/^{86}Sr$ plot comparing the compositions of Harappan potteries with that of the different types of Ghaggar flood plain sediments. It can be observed that the isotopic compositions of pottery samples lie within the range of brown silty-clay/surface mud and yellowish-brown sand. Possible contribution from surrounding aeolian sand cannot be ruled out. However, there appears to be a clear absence of any grey micaceous sand component within the pottery, which suggests non-availability of such sediment during pottery making. This on the other hand implies that by the time the Mature Harappans settled in Kalibangan, the glacial connection to the Ghaggar was significantly reduced and little sediment originating from glaciated terrains was depositing in the channels. Validation of this hypothesis comes from isotopic compositions of the brick sampled from the Bhatner Fort. It is a well-known historical fact that the Fort was established on the banks of an ephemeral Ghaggar during 12th century AD. The bricks of the fort, made using Ghaggar sediments, show similar compositions as that of the pre-historic potteries. This clearly suggests use of identical raw materials even after two millennia which in turn supports the theory that the river was already ephemeral (not glacier fed) during the Mature Harappan period.

5. Conclusion

The Ghaggar alluvium is a repository of sediments originated from two distinct provenances. The oldest grey micaceous sand (~70–4.3 ka) originated from the glaciated Higher Himalayas, thus, indicating a strong fluvial past of the river. The younger yellowish-brown sand and brown silty-clay (16–0 ka) were sourced from the Sub-Himalayan rocks, in particular from the Siwalik Group, Kasauli and Dagsahi Formations. Temporally overlapping depositions of sediments derived from both the Higher Himalayas and the Sub-Himalayas during the late Pleistocene and early Holocene point to the dwindling state of the river and gradual disappearance of its perennial water sources. The depositional ages of the topmost brown coloured silty-clay horizon confirm that the river had already lost its glacial sources and became an ephemeral foothill-fed river by 3 ka. The exact period of the hydrological changes has been narrowed down by the Nd—Sr isotopic fingerprinting of the Kalibangan potteries. This study suggests that the local sediments, which were most likely used for manufacturing the pottery, were not derived from the glacier covered parts of the Himalayas but were derived from the foothills of the

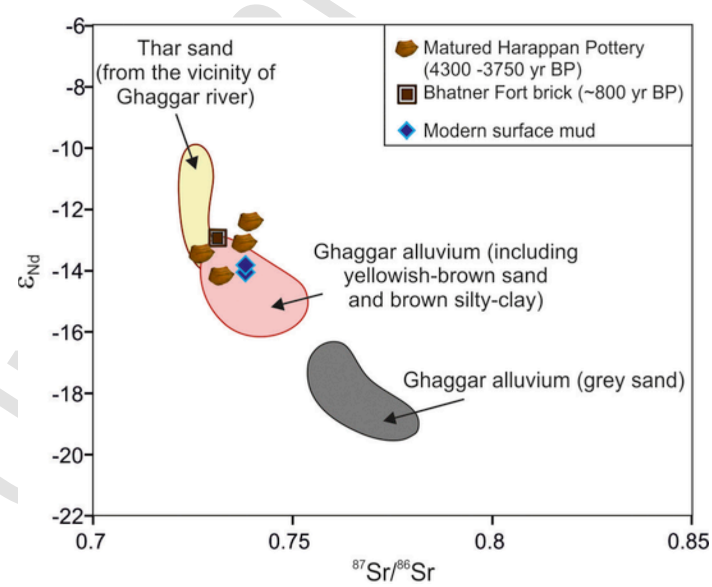

Figure 8. ε_{Nd} vs. $^{87}Sr/^{86}Sr$ plot of different archaeological artefacts compared with the probable raw material sources in the Ghaggar flood plain.

Himalayas. This in turn suggests that the Ghaggar had already transitioned from being a glacier-fed river to a rain-fed river during the Mature Harappan period.

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