Agricultural resilience and land-use from an Indus settlement in north-western India: Inferences from stable Carbon and Nitrogen isotopes of archaeobotanical remains

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Abstract

Stable isotopic compositions of carbon and nitrogen ($\delta^{13}C$, $\delta^{15}N$) of archaeological grains/seeds recovered from different cultural layers of an Indus archaeological site 4MSR (29°12'87.2"N; 73°9'421"E; Binjor, western Rajasthan, India) provide insights into the Harappan agriculture between ~ 2900 to ~ 1800 BCE. While $\delta^{13}C$ values were used to retrieve hydrological status, $\delta^{15}N$ values were used to gauge agricultural intensification. Isotopic data of grains/seeds were generated representing three Indus phases (i) early phase (~ 2900 – 2600 BCE), (ii) transitional phase (~ 2600 – 2500 BCE) and (iii) mature phase (~ 2500 – 1800 BCE). We find $\delta^{13}C$ values of barley grains (winter crop) varied in overlapping ranges for all the three phases − 21.3‰±1.9; −22.9‰±1.6 and − 22.7‰±1.7 respectively (*n* = 10 for each phase) indicating insignificant changes in hydrology for winter crops. For summer crops cotton also, average $\delta^{13}C$ values for transitional phase − 23.7‰±1.8 were not significantly different from those of mature phase − 22.5‰±2.4. $\delta^{15}N_{barley}$ values also varied in wider ranges, however, intra-phase variability appears to have progressively increased from early (8.0‰±1.7) to transitional phase (7.3‰±2.5) and then mature phase (9.2‰±7.2) indicating a plausible agricultural intensification. We also measured $\delta^{13}C$ of host soil organic matter (SOM) and sediment $\delta^{15}N$ to assess regional environmental conditions. In contrast to the trends observed for archaeological grains, $\delta^{13}C_{SOM}$ values showed a statistically significant enriching trend from early (−23.6‰±1.4) to mature phase (−20.3‰±1.9) hinting a growing aridity in the region. We surmise that Harappan farmers of western Rajasthan region might be managing arable hydrological conditions in their fields through agricultural interventions to continue agriculture practices despite growing aridity in the vicinity.

Introduction

Evidences of first organized life-style and agrarian human subsistence are found since the beginning of the Indus Valley Civilization that spread along the Indus and the Ghaggar-Hakra river systems (present day India-Pakistan international border) (Giosan et al., 2012; Levey and Burkey 1959; Possehl 2002; Weber et al., 2010). While human subsistence during the early phase was mainly dependent on agricultural and pastoral activities, however the mature phase witnessed variety of artisan activities and trade of goods with other contemporary civilizations (Park and Shinde, 2014). It has been argued that the Indus Civilization experienced a significant shifts in the environmental conditions *i.e.* arrival of monsoonal dryness during the mature phase which possibly led migrations and decline of this civilization (Dixit et al., 2014; Enzel et al., 1999; Giosan et al., 2012; Kathayat et al., 2017; Mac Donald, 2011; Petrie et al., 2017; Pokharia et al., 2017; Possehl, 2002; Sarkar et al., 2016; Sharma et al., 2020a; Singh et al., 1971; Staubwasser et al., 2003; Wright, 2010). Did these environmental changes affect agricultural strategies and production especially from early to mature phase of Indus era? While palaeo-climatologists broadly in agreement about monsoonal dryness prevailing during the latter part of the mature phase (Dixit et al., 2014; Giosan et al., 2012; 2018; Gupta et al., 2003; Kathayat et al., 2017; Mac Donald, 2011; Sarkar et al., 2016; Staubwasser et al., 2003 Wasson et al., 1984), archaeo-botanists continue to debate its impact(s) on agriculture (Petrie and Bates, 2017; Petrie et al., 2017; Pokharia et al., 2011, 2014, 2017; Sharma et al., 2020a; Singh et al., 1971, 1974). These studies yield two schools of thoughts: (i) monsoonal climate played a major role in shaping up the Harappan life-style, especially its subsistence (Bates et al., 2017; Kaushal et al, 2019; Petrie and Bates, 2017; Petrie et al., 2016;

Pokharia et al. 2011, 2014, 2017; Sharma et al., 2020a; Sarkar et al., 2016; Weber 2003) and (ii) Harappan farming ironically arrived during deteriorating monsoonal conditions and was adaptive in nature, since its inception. Geological records largely support the latter view (Berkelhammer et al., 2012; Dixit et al., 2014; Enzel et al., 1999; Giosan et al., 2012, 2018; Gupta et al. 2003; Kathayat et al., 2017; Mac Donald, 2011; Prasad and Enzel 2006; Staubwasser et al., 2003).

In this communication, we present stable isotopic C and N isotopic data of archaeological grains/seeds recovered from an archaeological site 4MSR of western Rajasthan (India) which tends to reinforce the latter hypothesis. In addition to these, we also measured C and N isotopes of soils derived from host habitational sediments. As the study site is a rural settlement and it can be well expected that archaeological/ habitational soil-sediment recovered from different cultural layers would have come from nearby arable land. Combined usage of C and N isotopes of crops vis-à-vis host soil-sediments of the studied archaeological sites was aimed to provide integrated information of cropping pattern and agro-hydrological status in arable Harappan agri-fields (Agnihotri et al., 2021; Wang et al., 2008). Where macro-botanical grains are expected to provide information about past crop-hydrological status, C and N isotopic data (along with their contents viz. TOC% and TN%) of soils recovered from different cultural periods may provide information about contemporary ecological status *i.e.* vegetation type (C-3 versus C-4) and in turn dryness of soils (Agnihotri et al., 2021; Araus et al., 1997; Ma et al., 2012; Parker et al., 2011; Peukert et al., 2012; Pokharia et al., 2017; Rosen et al., 1999; Simpson et al., 1999; Styring et al., 2016; Wang et al., 2008). This rationale is predominantly rely on the principle that C isotopic data is a good indicator of palaeo-hydrology and vegetation type (Ferrio et al., 2005; Ma et al., 2012; Parker et al., 2011; Wang et al., 2008)d isotopic data of plants/crops and soils can be governed by both environmental factors as well as agricultural amendments employed in agricultural fields (such as manuring, irrigation etc.) (Araus et al., 1997; Aguilera et al., 2008, 2017; Bogaard et al., 2013; Ferrio et al., 2005; Lee et al., 2005; Riehl et al., 2014; Styring et al., 2016, 2017; Wang et al., 2008; Wallace et al., 2013, 2015).

Different geological locations may have different sensitivities of these isotopic proxies (Jones et al., 2021 and reference therein), semi-arid conditions of the studied site 4MSR (Binjor) situated on the bank of dried Ghaggar river channel (~ 160 km away from major urban centre of Indus Civilization *i.e.* Harappa town) could be an ideal locale to investigate agricultural manifestations versus hydrological status of agri-fields in the past. For the 4MSR site, twelve (12) cultural layers were identified by archaeologists based on the material culture (such as pottery type, tools, seals etc.) and these cultural layers cover a total time span from ~ 2900 BCE to ~ 1800 BCE. Scientific chronologies of cultural layers were established by a combination of AMS and conventional radiocarbon dating of macro-botanical remains and soil organic matter recovered from nine strata (Sharma et al. 2020a and b).

It has to be noted that majority of the archaeological researches carried out so for on Indus valley sites provide wealthy information about their material-culture, advent of metal technologies, architectural prowess, trade, and overall socio-economic status of ancient settlers (Agarwal, 1971; Asthana, 1993; Bhan et al., 2002; Giosan et al., 2012; Kenoyer and Miller, 1999; Lal et al., 2003; Marshall, 1931; Possehl, 2002; Sana Ullah, 1931, 1940; Sharma et al., 2020b; Shinde, 2016; Vats, 1940). To the best of our knowledge, studies on Indus agronomy, crop-diversity, strategies and intensification are relatively sparse (García-Granero et al., 2016; Miller et al. 2006, 2015; Petrie and Bates, 2017; Petrie et al., 2016; Weber et al. 2010). This study provides first set of C and N isotopic data of Indus crops from a well dated archaeological site spanning the beginning of early phase to the end of mature phase. We also made an attempt to contextualize the derived palaeo-agricultural information with available knowledge gleaned from contemporary European and Chinese archaeological sites.

Chronology, archaeological background and macro-botanical details

The site 4MSR (29°12'87.2"N; 73°9'421"E), is situated at dry alluvial bed of Ghaggar river in the western Rajasthan (Fig. 1A). This area along the India-Pakistan border of western Rajasthan (District Anupgarh) is known to be a semi-arid region. Based on ^{14}C dating of charcoal, archaeological grains (barley, wheat and rice) and habitational soil-sediments, chronology of all the twelve cultural layers were ascertained (please see supplementary Table 1S). The total time-span covered by the obtained chronology has been sub-divided into

Figure 1 (A) Map showing location of archaeological study site 4MSR, Rajasthan along with other Indus sites (created by using mapping software *ArcGIS 10.3*). (B) Contour map of the study site shows excavated trenches and upper right shows the aerial view of the excavated site taken by dronne. (C) Section shows the location of sample collection from the mature Harappan and transitional phase stratigraphic layers. (D) Trench representing the sample collection area from the early Harappan phase deposit.

three time-windows following traditional Indus chronology (Kenoyer 1991). According to this convention, three periods are: the early phase ca. ~2900 – 2600 BCE; transitional phase ~ 2600 – 2500 BCE and the mature phase ~ 2500 – 1800 BCE. Figure 1B, contour map of the site shows excavated trenches with stratigraphic details from natural bed sediment to modern humus i.e. decreasing depth in mean sea level unit. Section in the Fig. 1C and D show the location of soil-sediment sample collection from the early, transitional and mature phase stratigraphic cultural layers. As stated earlier, there are twelve distinct identified cultural layers. The upper layers (one to four) stored remnants of mature phase, while middle layers (five to seven) represented a transitional phase followed by deeper cultural layers belonging to the early phase of the Indus era.

Several studies have been conducted along the palaeo-channel of Ghaggar (erstwhile Saraswati) river, originating from northwest Himalayas and supposed to have flown southwest towards the Gujarat Kachchh region (Chatterjee et al., 2019; Ghose et al., 1979; Giosan et al., 2012; Gupta et al., 2004; Joshi et al., 1984; Kar et el., 2004; Lal, 2002; Marshall, 1931; Oldham, 1886, 1893; Possehl, 1999; Singh et al., 2017; Yashpal et al., 1980). This relict channel is thought to have catered several pre-historic phases of human civilizations of Indus culture in the north-western India (Giosan et al., 2012; Lal et al., 2003; Marshall, 1931; Mughal, 1997; Possehl, 2002, Sharma et al., 2020a; Vats, 1940 and references therein). The site 4MSR (locally known as Binjor) evolved from a typical agricultural settlement to a major rural industrial centre that manufactured copper artefacts, beads from semi-precious stones and a wide variety of terracotta products and may have exported them to other urban Harappan sites nearby and far-flung areas. A large series of different shapes of hearths, furnaces and kilns with a cluster of multi-purpose workshops for industrial activities indicate a rural metal-factory settlement at the site 4MSR (Sharma et al., 2020b). A range of recovered artifacts of gold and copper, seashells and terracotta such as pendant frames, earrings, beads, spacers, chisels, bangles, needles, fish hooks, big storage pots, twin pots, broken perforated jars, terracotta beads and broken terracotta bangles, weights, seals, terracotta toys (humped bulls) confirmed the industrial nature of the site (Sharma et al., 2020b).

The macro-botanical assemblage recovered from the site is shown in supplementary Fig. 1S (data adopted from Sharma et al., 2020a). The assemblage was comprising of variety of cereals and leguminous crops *viz*., *Hordeum vulgare*, *Triticum aestivum/ durum*, *Oryza sativa*, *Setaria* sp., *Pisum arvense*, *Lens culinaris*, *Cicer arietinum*, *Lathyrus* sp. and *Vigna* sp. etc. Besides these, oleiferous and fibrous crops *viz. Sesamum indicum*, *Linum usitatissimum* and *Gossypium* sp. were also been recorded (Sharma et al., 2020a). Figure 2 (data adopted from Sharma et al., 2020a) presents pie charts depicting relative proportions of Indus summer (*Oryza sativa*, *Setaria* sp., *Vigna* sp., *Gossypium* Sp. and *Sesamum indicum*) and winter (*Hordeum vulgare*, *Triticum* sp., *Pisum arvense*, *Lens culinaris*, *Cicer arietinum*, *Lathyrus* sp. and *Linum usitatissimum*) crop species during early, transitional and the mature phase. It is noteworthy here that summer crops appear to have introduced mainly during the transitional phase (Fig. 2B). Intensification and diversification of various crops appear to dominate Indus croplands belonging to the mature phase (Fig. 2C). The 4MSR archaeological site has yielded various metal artifacts along with variety of domestic hearths from the cultural layers belonging to the mature phase of Indus era (Sharma et al., 2020a and b).

Materials and Methods

Macro-botanical remains comprise plant traces that are large enough to be recognized with the naked eye or low-powered microscope (Ford 1979; Fritz 2005; Pearsall 2000). The macro-botanical samples (archaeological grains/seeds; supplementary Fig. 1S) were collected from all the excavated trenches. Major collection areas were floor, hearths and pits (belonging to the mature phase; from topmost cultural layer number 1 to number 4), soils of the transitional phase (layer number 5 to 7) and early phase (deeper cultural layers number 8 to 12). Botanical grains/seeds were separated from soil using water floatation technique in the field itself (Sharma et al., 2020a). Segregated samples were identified as per taxonomical classification up to the genus and species level and photo-documented (Sharma et al., 2020a). For carbon and nitrogen stable isotopic measurements, segregated archaeological grains of barley (winter crop; n = 10), seeds of cotton (summer crop; n = 10) and tiny vetch (leguminous weed; n = 10) were selected following criteria of the preservation class three (Hubbard and Al Azm, 1990). Binning of grains /seeds was done to represent different phases of Indus era *i.e.* early, transitional and mature phase. It is noteworthy that even though the study site falls under semi-arid region of western Rajasthan (District Anupgarh), modern day landscape appears to be significantly influenced by Indira canal in the vicinity (since 1980s) which changed arable fields in to lush green farm lands (Fig. 1B) (Anonymous, 2021).

We also collected soil-sediments from all the twelve stratigraphic cultural layers along with the natural bed sediment (beneath the oldest (bottommost) cultural layer of early phase) and sediment belonging to modern humus from the vicinity of the archaeological site for measuring their C and N isotopic compositions. The site 4MSR is situated at dry alluvial bed of Ghaggar river (Ali, 1941; Oldham, 1893) which is now extinct.

During its active phase, the channel was receiving waters and alluvium from the Himalaya (Kar, 2011; 2014). The alluvium of this dry bed is of variable texture mainly representing from sandy loam to clayey sediments (Shyampura and Sehgal 1995). The surface soil in the area is fertile for agriculture (Anonymous, 2021).

Carbon and Nitrogen isotopic analyses of archaeological grains/ seeds and soil-sediments

We measured the $\delta^{13}C$ and $\delta^{15}N$ values along with C and N contents of archaeological grains/seeds (and soil-sediments) using an Elemental Analyzer (EA; Pyrocube®, Elementar® Germany) coupled with Isotope Ratio Mass spectrometry (EA-IRMS; Precision®, Elementar® UK) in continuous flow mode. A total of seventy nine (79) archaeological grains/seeds (of barley, cotton, tiny vetch, pulses, oil and fibre crops) were collected from the different cultural layers representing all the three phases (early, transitional and mature phase). The archaeological grain/seed samples were mildly washed to remove any adhering dirt/soil because these samples were collected by the water floatation technique which has already removed the soil from the grain/seed samples. The washed grains/seeds were then dried at 60°C for 48 hours and then stored in vials. The dried grain samples were mildly powdered using an agate mortar and pestle. ~0.2-1.0 mg weights of these were taken in clean tin cups, which were subsequently pressed in to oval shaped pellets. Hence, due to sandy loam nature of soil and ease with which it was cleaned from grain/seed samples, no pre-chemical treatment was carried out to further clean them (such as etching). We followed the analysis with mild cleaning and without pre-treatment to avoid any altertion in the original isotopic compositions. It is based on the literature survey of several studies that suggested $\delta^{13}C$ values in cereal grains are not significantly affected by carbonisation within the range of temperatures between ~ ~ 200–400°C) (Araus et al., 1997; Aguilera et al., 2008; Ferrio et al., 2007). There are studies (Charles et al., 2015; Fraser et al., 2013; Kaushal et al., 2019) that have demonstrated that lower charring temperatures between 200–240°C (for about a duration of six hours) under reducing conditions produce morphologically intact carbonized grains. For $\delta^{15}N$ signal also, Bogaard et al. (2007) reported insignificant changes for grains carbonized at 230°C for up to 24 hours. Therefore, we chose morphologically intact (well-preserved) archaeological carbonized grains/seeds only upto the preservation class three (Hubbard and Al Azm, 1990) for isotopic analyses. It is important to underscore that earlier archaeo-isotope workers have not catagorically reported any significant differences between pre-treated and non-treated grains samples (Aguilera et al. 2017; Kaushal et al., 2019; Lightfoot and Stevens 2012; Styring et al. 2016;).

Samples of host soil-sediments, however, were pre-treated *i.e.* subjected to decalcification to measure the $\delta^{13}C_{SOM}$ values (Agnihotri et al., 2021). For this, ~ 1–2 grams of dried/homogenized sediment aliquots (without sieving to avoid biasness) were treated with 5% Hydrochloric acid and kept for ~ 8–10 hours (overnight) at room temperature. Treated samples were then washed several times with deionised water to remove any excess of chloride ion. Washed sediment samples were then again dried in an oven at ~ 50–60°C followed by mildly re-powdering with a pestle in an agate mortar. Finally grounded powders were dried and dried powders were then transferred in clean plastic vials. A total of 51 samples were analysed on EA-IRMS. Data quality of measured C and N contents together with $\delta^{13}C$ and $\delta^{15}N$ values was checked throughout the analysis using a suite of *in-house* and international IAEA standards. Accuracy of measured

$\delta^{13}C$ and $\delta^{15}N$ data was better than 0.2‰, and for elemental concentrations, it was better than 5% (based on duplicate analysis). Detailed methodology of the used EA-IRMS facility is published elsewhere (Agnihotri et al., 2020). The isotopic data are reported using the standard delta notations. Standard for determining C isotopic data is Vienna-PDB, while N isotopic data is normalized with $^{15}N/^{14}N$ ratio of atmospheric N_2 (Tables 1 and 2). Chemical and isotopic data are arranged phase wise, constrained by cultural identification of habitational layers (based on materials such as pottery, seal, metal objects etc.) and the determined radiocarbon chronology (Sharma et al., 2020a; Tables 1 and 2).

Estimation of carbon isotope discrimination factor ($\Delta^{13}C$)

We computed the $\Delta^{13}C$ values using measured $\delta^{13}C$ values of crop grains/seeds to evaluate crop-water status (Aguilera et al., 2008; 2012; Farquhar et al., 1989; Ferrio et al., 2005; Styring et al., 2016; Wang et al., 2008). $\Delta^{13}C$ values were computed following Farquhar et al. (1989) using the formula-

$\Delta^{13}C = (\delta^{13}C_{air} - \delta^{13}C_{plants}) / (1 + \delta^{13}C_{plant}/1000)$——————(1)

Past $\delta^{13}C_{air}$ values were inferred by interpolating a range of data from Antarctic ice-core records together with modern data from two Antarctic stations (Halley Bay and Palmer Station) of the CU-INSTAAR/NOAA-CMDL network for atmospheric CO2 (ftp://ftp.cmdl.noaa.gov/ccg/co2c13/flask/readme.html). In fact, slightly different $\delta^{13}C_{air}$ values for the early and mature Harappan phase (−6.3‰ and −6.4‰ respectively) were used. We used estimated $\Delta^{13}C_{crop\text{-}remains}$ values to ascertain crop-water status (Araus et al., 1997; Aguilera et al., 2008, 2017; Ferrio et al., 2005; Lee et al., 2005; Riehl et al., 2014; Styring et al., 2016, 2017; Wang et al., 2008; Wallace et al., 2013, 2015).

Indus landscapes were resource-rich (in terms of availability of fertile soil and seasonal rainfall in both summer and winters), numerous archaeological evidences reveal Harappan settlers were using water management practices in dwelling areas as well as in farming (by channelling of river water to crop fields or using ground waters for irrigation purpose). For instance, Harappans apparently evolved a more developed irrigation technology than their predecessors that allowed to exploit the spacious and fertile Indus River basin (Kenoyer 1991b). The network of dams, canals and reservoirs at Indus/ Harappans site Dholavira indicates elaborated water management system during the Indus time (Bisht 1998–1999). Hence, human efforts might have played a significant role to manage agriculture activities required to feed communities. Several recent studies attempted to glean clues about changes in past agronomy under the changing monsoonal climate (Pokharia et al., 2017; Sarkar et al., 2016; Sharma et al., 2020a). These studies remained devoid of any numerical data to compare hydrological status of Indus farmlands.

Statistical analyses

We conducted non-parametric ‘One way ANOVA test (Kruskal-Wallis test) which investigates population of data categorized among different phases by retaining or rejecting the null hypothesis. These statistical validation tests were performed for both C and N isotopic data generated for grains/seeds as well as sedimentary layers using SPSS software (version# 21).

Results

Measured $\delta^{13}C$ and $\delta^{15}N$ values along with the estimated $\Delta^{13}C$ values of archaeological grains/seeds *viz.* barley, cotton and tiny vetch from all the three phases (early, transitional and mature phase) have been presented in the Table 1. Tiny vetch is an associated leguminous weed of winter crops like barley, wheat, gram and lentil (Zohaib et al., 2014). Crop-water status of winter and summer seasons were assessed using $\Delta^{13}C$ values of barley and the cotton crops respectively. Measured $\delta^{13}C_{SOM\ and\ TC}$, $\delta^{15}N_{TN}$, Total Organic Content/Total Nitrogen (TOC/TN) or simply C/N weight ratios, Total Inorganic Content/Total Organic Content (TIC/TOC) values of soil-sediments from different cultural (occupational) phases have been presented in the Table 2.

Table 1

Measured δ[13]C and δ[15]N values along with the estimated Δ[13]C values of archaeological barley, cotton and common/tiny vetch (weed) grains/ seeds. Total number of grains/ seeds *n* = 10 has been selected for each crop/weed from all the three phases each phase). (Abbreviations: Total Nitrogen (TN%), Total Carbon (TC%), Carbon/Nitrogen (C/N weight ratios).

Sample ID	**TN (%) (Measured)**	**$\delta^{15}N$ (‰) (Measured)**	**TC (%) (Measured)**	**$\delta^{13}C$ (‰) (Measured)**	**$\Delta^{13}C$ (‰) (Estimated)**	**C/N (Calculated)**
***Hordeum vulgare* (Barley)**						
Early Harappan Phase						
BE-1	1.9	7.9	47.5	-21.5	15.5	24.5
BE-2	1.7	8.4	43.8	-18.9	12.8	26.3
BE-3	1.8	8.1	46.9	-21.2	15.2	25.5
BE-4	1.2	8.4	41.4	-21.3	15.3	35.5
BE-5	2.0	6.4	49.1	-20.5	14.5	24.9
BE-6	3.1	9.2	43.9	-18.3	12.3	14.2
BE-7	1.5	11.0	42.8	-22.2	16.2	28.3
BE-8	1.9	7.0	46.4	-21.2	15.2	24.1
BE-9	0.9	4.8	38.4	-24.5	18.7	41.5
BE-10	2.1	5.8	40.4	-23.9	18.0	19.7
Transitional Phase						
BT-1	1.7	6.6	43.5	-23.9	18.0	25.0
BT-2	2.7	7.3	51.9	-21.6	15.6	19.2
BT-3	1.5	8.4	45.0	-24.4	18.6	29.4
BT-4	1.6	12.8	46.6	-22.6	16.7	30.0
BT-5	2.5	12.7	45.6	-20.0	14.0	18.0
BT-6	4.0	5.3	43.1	-19.7	13.7	10.7
BT-7	2.1	6.7	45.0	-23.1	17.2	21.5
BT-8	2.2	7.5	46.0	-22.6	16.7	20.5
BT-9	2.0	7.3	43.4	-24.1	18.3	21.3
BT-10	3.0	6.9	43.0	-23.4	17.5	14.5

Sample ID	TN (%) (Measured)	$\delta^{15}N$ (‰) (Measured)	TC (%) (Measured)	$\delta^{13}C$ (‰) (Measured)	$\Delta^{13}C$ (‰) (Estimated)	C/N (Calculated)
Hordeum vulgare (Barley)						
			Mature Harappan Phase			
BM-1	3.2	4.9	48.4	-20.0	13.9	15.3
BM-2	1.9	7.7	43.4	-21.6	15.5	22.3
BM-3	2.8	13.3	43.2	-21.9	15.8	15.5
BM-4	2.0	6.1	41.5	-24.3	18.3	20.7
BM-5	1.7	30.3	44.7	-24.1	18.2	26.3
BM-6	2.5	10.4	48.7	-23.2	17.2	19.8
BM-7	2.5	8.0	55.0	-23.7	17.7	22.3
BM-8	3.0	9.0	48.2	-25.5	19.6	15.9
BM-9	1.8	12.6	47.6	-22.3	16.2	26.5
BM-10	2.9	9.4	39.7	-21.0	14.9	13.8
Viciasp. (common/tiny vetch, weed)						
Transitional Phase						
VT-1	4.2	4.7	44.9	-21.6	15.6	10.7
VT-2	5.5	5.6	43.5	-20.0	14.0	7.9
VT-3	3.9	1.0	40.3	-22.5	16.5	10.3
VT-4	5.0	1.6	43.9	-24.5	18.7	8.8
VT-5	4.4	10.5	44.1	-23.7	17.8	9.9
VT-6	5.2	4.1	44.7	-22.5	16.6	8.6
VT-7	5.3	4.3	40.1	-22.4	16.4	7.5
VT-8	5.0	1.6	40.8	-23.7	17.9	8.2
VT-9	4.5	5.9	40.6	-24.7	18.9	9.0
VT-10	4.8	2.6	37.6	-23.4	17.5	7.8
Mature Harappan Phase						
VM-1	1.9	1.8	18.3	-23.8	17.8	9.7

Sample ID	TN (%) (Measured)	$\delta^{15}N$ (‰) (Measured)	TC (%) (Measured)	$\delta^{13}C$ (‰) (Measured)	$\Delta^{13}C$ (‰) (Estimated)	C/N (Calculated)
Hordeum vulgare (Barley)						
VM-2	3.5	2.6	46.9	-24.1	18.1	13.2
VM-3	4.2	0.8	42.7	-24.2	18.2	10.1
VM-4	4.8	0.8	41.8	-26.1	20.3	8.7
VM-5	4.7	1.1	41.4	-24.7	18.8	8.8
VM-6	6.0	7.1	42.0	-24.4	18.5	7.0
VM-7	4.0	1.3	44.5	-24.7	18.8	11.2
VM-8	3.4	3.1	36.3	-21.4	15.3	10.6
VM-9	4.0	3.6	40.4	-23.3	17.3	10.0
VM-10	3.9	4.5	31.8	-22.9	16.9	8.1
Gossypiumsp. (Cotton)						
Transitional Phase						
GT-1	4.3	1.2	45.8	-22.7	16.7	10.6
GT-2	4.6	12.3	36.4	-23.0	17.1	8.0
GT-3	4.6	20.5	32.8	-24.2	18.3	7.1
GT-4	5.2	5.1	39.4	-23.3	17.4	7.5
GT-5	5.4	22.1	43.7	-24.5	18.6	8.2
GT-6	2.1	4.2	47.0	-25.1	19.3	22.4
GT-7	3.7	14.1	31.5	-20.5	14.5	8.4
GT-8	4.2	1.3	42.7	-24.8	19.0	10.2
GT-9	4.7		43.8	-20.5	14.5	9.3
GT-10	2.0	11.9	52.3	-25.8	20.0	25.6
Mature Harappan Phase						
GM-1	3.6	12.9	45.6	-25.1	19.2	12.5
GM-2	4.6	16.4	38.2	-20.1	13.9	8.3
GM-3	5.2	9.4	34.8	-23.1	17.1	6.7
GM-4	5.4	7.1	37.0	-23.8	17.9	6.8

Sample ID	TN (%) (Measured)	$\delta^{15}N$ (‰) (Measured)	TC (%) (Measured)	$\delta^{13}C$ (‰) (Measured)	$\Delta^{13}C$ (‰) (Estimated)	C/N (Calculated)
Hordeum vulgare (Barley)						
GM-5	4.5	15.7	35.3	-20.0	13.9	7.8
GM-6	4.5	9.3	35.5	-22.0	16.0	7.9
GM-7	5.0	13.7	36.9	-19.1	13.0	7.3
GM-8	4.2	5.4	34.4	-22.4	16.3	8.2
GM-9	4.3	10.7	35.8	-22.6	16.6	8.3
GM-10	1.1	9.5	45.5	-27.2	21.4	39.9

Table 2

Measured $\delta^{13}C_{SOM\ and\ TC}$, TC%, $\delta^{15}N_{TN}$ and TN% values along with calculated TOC/TN, TIC/TOC ratios of soil-sediments from different occupational phases at 4MSR, Rajasthan, India. (Abbreviations: SOM = Soil Organic Matter, TC = Total Carbon, TOC = Total Organic Carbon, TN = Total Nitrogen, TIC = Total Inorganic Carbon).

Sample ID	TN (mg/g)	$\delta^{15}N$ (‰)	TC (mg/g) bulk	$\delta^{13}C$ (bulk) (‰)	$\delta^{13}C$ (SOM) (‰)	TOC (mg/g)	TIC (mg/g)	TOC/TN	TIC/TOC
Humus (Modern) and Agricultural (wheat) field Sediment									
S1	26.4	7.3	549.4	-12.4	-25.2	159.3	390.1	6.0	2.4
S2	55.8	8.7	741.0	-9.5	-24.3	376.4	364.6	6.7	1.0
S3	41.2	7.6	851.7	-12.5	-25.3	597.7	254.0	14.5	0.4
S4	135.9	7.5	960.7	-15.6	-25.1	266.0	694.7	2.0	2.6
S5	47.1	7.4	674.5	-16.1	-25.8	289.5	385.0	6.1	1.3
S6	14.9	9.4	527.4	-6.6	-22.1	270.0	257.3	18.2	1.0
AF	40.1	8.5	684.0	-11.7	-23.5	153.6	530.4	3.8	3.5
Mature Harappan and Transitional Phase Sediment									
MH1a	42.6	8.7	875.4	-9.2	-18.6	580.0	295.4	13.6	0.5
MH1b	34.7	8.3	802.8	-6.1	-19.6	310.0	492.8	8.9	1.6
MH2	14.0	11.5	526.4	-7.1	-19.8	260.0	266.4	18.5	1.0
MH3a	30.5	14.0	767.7	-9.8	-21.4	415.0	352.7	13.6	0.8
MH3ax	15.9	14.0	892.4	-12.1	-20.9	310.0	582.4	19.4	1.9
MH3ay	2.5	16.2	219.6	-4.9	-23.2	80.0	139.6	31.7	1.7
MH3az	3.9	15.7	240.6	-6.1	-23.3	100.0	140.6	25.4	1.4
MH3b	18.9	11.0	605.3	-9.8	-21.1	280.0	325.3	14.8	1.2
MH4a	32.6	9.9	680.5	-8.1	-18.0	420.0	260.5	12.9	0.6
MH4b	35.4	10.5	1028.8	-12.1	-18.2	820.0	208.8	23.2	0.3
TP5	42.6	9.7	1222.3	-13.3	-22.5	620.0	602.3	14.6	1.0
TP6	25.0	8.5	870.0	-8.0	-22.8	370.9	499.1	14.8	1.3
Early Harappan Phase Sediment									
EH1	17.3	10.5	1359.2	-11.0	-24.2	305.8	1053.3	17.7	3.4
EH2	13.9	9.8	1861.4	-10.9	-23.5	383.4	1477.9	27.6	3.9

Sample ID	TN (mg/g)	$\delta^{15}N$ (‰)	TC (mg/g) bulk	$\delta^{13}C$ (bulk) (‰)	$\delta^{13}C$ (SOM) (‰)	TOC (mg/g)	TIC (mg/g)	TOC/TN	TIC/TOC
EH3	22.0	10.9	1207.8	-10.4	-24.5	521.8	686.0	23.7	1.3
EH4	23.0	9.7	765.2	-7.3	-23.2	248.0	517.2	10.8	2.1
EH5	16.7	7.7	602.7	-5.1	-24.0	122.4	480.3	7.3	3.9
EH6	10.7	6.9	545.2	-4.7	-22.9	174.2	371.1	16.2	2.1
EH7	9.6	14.5	559.0	-4.8	-25.1	113.6	445.5	11.8	3.9
EH8	17.4	9.2	840.8	-9.9	-23.1	378.6	462.2	21.8	1.2
EH9	15.9	14.1	716.8	-5.9	-24.4	153.3	563.5	9.7	3.7
EH10	26.7	7.8	740.6	-7.7	-23.1	286.4	454.2	10.7	1.6
EH11	15.6	7.3	658.6	-5.7	-24.8	108.0	550.6	6.9	5.1
EH12	16.5	6.8	868.0	-7.1	-23.8	164.3	703.7	10.0	4.3
EH13	33.5	6.7	893.2	-9.9	-19.2	462.0	431.1	13.8	0.9
EH14	17.0	8.1	1028.2	-7.4	-23.1	286.9	741.3	16.9	2.6
EH15	11.7	11.6	782.5	-7.4	-23.1	135.1	647.4	11.6	4.8
EH16	9.2	8.2	599.9	-3.2	-23.6	136.7	463.2	14.9	3.4
EH17	8.5	14.3	1266.7	-3.1	-24.1	91.6	1175.2	10.7	12.8
EH18	13.1	5.1	528.3	-3.2	-23.9	165.2	363.1	12.6	2.2
EH19	13.4	6.7	543.9	-3.4	-24.2	103.7	440.1	7.7	4.2
EH20	13.9	7.9	572.4	-3.7	-23.7	114.1	458.3	8.2	4.0
EH21	12.9	12.9	559.1	-2.8	-22.6	103.8	455.3	8.0	4.4
EH22	12.9	7.4	550.8	-4.9	-21.7	152.0	398.8	11.8	2.6
EH23	17.7	10.5	485.0	-3.6	-21.6	116.7	368.3	6.6	3.2
EH24	15.5	9.7	464.0	-3.9	-22.1	136.1	327.8	8.8	2.4
EH25	14.9	11.0	435.1	-3.3	-23.3	103.2	331.8	6.9	3.2
EH26	7.1	7.5	505.3	-3.3	-23.3	90.3	415.1	12.7	4.6
EH27	8.9	6.5	543.1	-3.4	-27.2	84.9	458.1	9.6	5.4
EH28	5.1	7.6	488.5	-3.0	-25.1	54.5	434.0	10.8	8.0
EH29	12.1	6.9	520.6	-3.3	-24.1	131.1	389.5	10.9	3.0

Sample ID	TN (mg/g)	$\delta^{15}N$ (‰)	TC (mg/g) bulk	$\delta^{13}C$ (bulk) (‰)	$\delta^{13}C$ (SOM) (‰)	TOC (mg/g)	TIC (mg/g)	TOC/TN	TIC/TOC
Bed Sediment									
BS1	1.9	-3.4	529.1	-3.0	-27.7	147.6	381.4	79.5	2.6
BS2	5.6	7.4	566.3	-3.0	-26.2	93.8	472.5	16.8	5.0
BS3	6.1	12.2	659.0	-3.2	-24.0	126.9	532.1	20.8	4.2

Figure 3 shows the box-whisker plots of the measured $\delta^{13}C$ values of archaeological grains of barley and seeds of cotton and weed (tiny vetch) during the early, transitional and mature phase. Average $\delta^{13}C$ values of barley for the early, transitional and mature phase were found to be −21.3‰±1.9, −22.9‰±1.6 and −22.7‰±1.7 respectively (Fig. 3(A); Table 1). Thus they were found to have varied in overlapping ranges which can be interpreted in terms of similar hydrological conditions throughout, albeit a marginally drier status for the grains of the early phase. Summer crops appeared mainly during the transitional phase (Sharma et al., 2020a) and average $\delta^{13}C_{cotton}$ values during the transitional phase (−23.7‰±1.8) were found to be marginally depleted compared to those during the mature phase −22.5‰±2.4, indicating better hydrological status of the transitional phase (with respect to those in mature phase) (Fig. 3(G). No significant differences were found for the $\delta^{13}C$ values for both barley and cotton during all the three identified phases. Statistical validations of the aforesaid statements were ascertained using non-parametric one way ANOVA tests which retained the null hypothesis.

The $\delta^{13}C$ values of the weed plant (not sown/ seeded crop) 'tiny vetch' were also used to gauge environmental conditions of Harappan agricultural fields. Average $\delta^{13}C_{tiny\ vetch}$ values for the transitional and mature phase were−23.0‰±1.4 and −24.1‰±1.2 respectively (Fig. 3(D); Table 1). These values also indicated more or less similar hydrological status during transitional and mature phase as yielded by winter crop barley (Fig. 3(A).

Figure 4A shows box-whisker plots of measured $\delta^{13}C$ values of the soil organic matter along with sediment $\delta^{15}N$ values and C/N weight ratios from different cultural phases of the same occupation during Indus era (Fig. 4B, C). C/N ratios of soils primarily indicate nature of soil, but they could also mimic environmental dryness *i.e.* higher the C/N ratio drier the soil is (Jiao et al. 2016). Average $\delta^{13}C$ values of soil organic matter during the early, transitional and mature phase (−23.6‰±1.4, −22.6‰±0.3, −20.3‰±1.9, respectively) depict a conspicuous enhancing trend from early to mature phase (Fig. 4A; Table 2). Statistical validation of aforesaid trend and distinctiveness of $\delta^{13}C$ values of soil organic matter during different phases (early, transitional and mature phase) was ascertained by the non-parametric One way ANOVA test which rejected the null hypothesis ($p = 0.000$). This enhancing trend could be interpreted as prevailing aridity in the region and it is well supported by $\delta^{15}N_{soil-sediment}$ values which also show an increasing trend (validated by one way ANOVA test; $p = 0.015$) from early (8.1‰±2.5) to mature phase (11.2‰±2.8) via transitional phase (9.1‰ ±0.9) (Fig. 4B; Table 2).

This increasing trend in $\delta^{15}N_{soil-sediment}$ could, however, may be due to complex mixture of environmental and agricultural intensifications (crop rotation and manuring activities) (Aguilera et al., 2008; Boggard et al. 2007, 2013; Bol et al. 2005; Choi et al. 2006; Fraser et al., 2011; Senbayram et al. 2008; Styring et al., 2016, 2019; Szpak et al., 2014). To investigate various possibilities, $\delta^{15}N$ values of archaeological grains were used in tandem with TN contents. Figure 3(B) displays $\delta^{15}N_{barley}$ values during early, transitional and mature phase (8.0‰±1.7, 7.3‰±2.5, 9.2‰±7.2 respectively) but fall within overlapping ranges (validated by one way ANOVA test; $p = 0.196$). It could be, however, noticed that intra-phase variability in $\delta^{15}N_{barley}$ values (depicted by standard deviations) increased from early to transitional and then mature phase. Total Nitrogen contents of barley grains showed overlapping ranges from the early (1.9%±0.6) to transitional (2.2%±0.8) and then mature phase (2.5%±0.5) (Table 1). The enhancing intra-phase variability of $\delta^{15}N_{barley}$ values from early to mature phase may be a combined effect of change in cropping pattern and agricultural intensification adopted by Harappan farmers.

For summer crop, $\delta^{15}N_{cotton}$ values during the transitional phase show much larger variability (11.9‰±7.8) compared to that of mature phase 10.1‰±3.6 most likely due to crop-diversification adopted during transitional phase and its impact on arable soil (Fig. 3(H); Table 1). However, $\delta^{15}N_{cotton}$ values did not show any statistically significant difference (validated by one way ANOVA test; $p = 0.799$) in early, transitional and mature phase ($p = 0.450$). Average TN contents of cotton crops also do not show significant variation (validated by one way ANOVA test; $p = 0.768$) from transitional phase to mature phase (4.4%±1.2 in transitional phase and 4.5%±1.2 in mature phase; Table 1). The weed plant (tiny vetch) also showed a larger degree of variability in $\delta^{15}N$ during the transitional phase (4.2‰±2.8) compared to that for mature phase (2.2‰±1.9) (Fig. 3(E); Table 1). Besides, tiny vetch showed the lower $\delta^{15}N$ values as expected for the leguminous plants (Fig. 3(E); Table 1). C/N values for archaeological barley grains shown in Fig. 3(C) indicate better degree of preservation for grains of the transitional and mature phase compared to those from early phase. Two higher (outlier) values for instance, clearly due to poorer preservation. Similarly the C/N ratios of tiny vetch and cotton seeds also showed the good degree of preservation (Fig. 3(F, I).

For comparing hydrological status of Harappan crops grown at the site 4MSR with hydrological status of crops (isotopic data) from other archaeological sites of the world, we used estimated $\Delta^{13}C$ values of barley, cotton and tiny vetch during different phases (Table 1). Estimated average $\Delta^{13}C$ values for barley from our study site were 15.3‰±2.0, 16.9‰±1.7, 16.7‰±1.8 for early, transitional and mature phase respectively (Table 1). Intriguingly, values during the early phase were found to be of drier hydrological status compared to those of transitional and mature phase. For summer crop cotton, average $\Delta^{13}C$ values for the transitional phase (17.9‰±1.9) indicated slightly wetter status compared to that of mature phase (16.5‰±2.6) (Table 1). In contrast, average $\Delta^{13}C$ values of tiny vetch (weed) indicated a drier hydrological status of this winter crop for the transitional phase compared to that of mature phase (17.1‰±1.5 and 18.2‰±1.3 respectively) (Table 1).

Discussion

Average $\delta^{13}C$ values of soil organic matter indicated a growing aridity from the early to mature phase (Fig. 4A), but crop archaeological seeds/grains grown during both winter and summer seasons (Fig.s 3(A) and (G) do not show such aridity trend. Our earlier work from an archaeological site Khirsara (23º27'N, 69º03'E) from a semi-arid region of southern Gujarat also demonstrated a major shift in crop-assemblage (towards drought resistant millet based crops) well supported by a significant enhancement in the $\delta^{13}C$ values of soil organic matter at ~ 2250 BCE (Pokharia et al., 2017). Cultural continuity was maintained despite this significant crop-shift likely enforced by prevailed monsoonal aridity. Several regional geological records do present evidences for significant monsoonal dryness (Dixit et al., 2014, 2018; Prasad et al., 2014; Staubwasser et al. 2003).

Enriched trend in $\delta^{13}C_{SOM}$ values (Fig. 4A) indicate environmental conditions possibly varied from wetter to drier conditions. This inference could be supported by macro-botanical assemblage that shows arrival of drought resistant crops (*e.g.* millets) during the late mature phase (Sharma et al., 2020a). Earlier, geological repositories of north-western India also have shown gripping aridity during the progressively increasing industrial (metallurgical) activities of Harappans based on the recovered material-culture (Dixit et al., 2014; Kathayat et al., 2017; Sarkar et al., 2016; Sharma et al., 2020b). Taken together, it appears that there was a gradual transformation in agriculture prowess of Harappan farmers from the early to mature phase which appears to have outplayed prevailing aridity in the region. All these evidences collaterally suggest agriculture continued to the end of the mature phase (~ 1800 BCE) together with upcoming industrial activities. $\delta^{13}C$ and $\delta^{15}N$ data of studied (well preserved) archaeological grains, however, do not show any significant changes. This observation lead us to infer that it is likely that Harappan farmers of western Rajasthan area were able to manage their agriculture by application of other means such as crop shifting, irrigation and alike interventions. More intrusive analyses of Indus agronomical treasures are needed. Drawn inferences, however, appear to be well corroborated by analysis of botanical assemblage from the site which noticeably shows (i) crop-diversification during intermediate transitional phase and (ii) advent of millet based crops during the latter part of mature phase, attributable to prevailed monsoonal aridity increased in the region (Sharma et al., 2020a).

Routson et al., (2019) have noted a decreasing trend in net precipitation during early to mid-Holocene globally (for mid-latitudes). This inference is well corroborated by regional hydro-climatic records of the Indian monsoon (Flietmann et al., 2003; Gupta et al., 2003). It has been demonstrated that the $\Delta^{13}C$ values of archaeological agricultural grains (such as barley) recovered from the different archaeological sites (along with the sites falling under semi-arid to arid zones) typically ranging between 15.0 to 22.0‰ were grown under ~ 100–250 mm of mean annual precipitation (Araus et al., 1997; Aguilera et al.2008; Ferrio et al., 2005; Gron et al., 2017, 2021; Kanstrup et al., 2014; Wallace et al., 2013, 2015; Vaiglova et al., 2020). Araus et al. (1997) have shown lower $\Delta^{13}C_{barley}$ values from archaeological sites in the Fertile Crescent and western Mediterranean (Spain) as indicator of poorer water status during arid epochs. Numerous other studies also have demonstrated that the lower $\Delta^{13}C$ values resulting from the poor to moderate water availability reflected by the grains grown in drier conditions (Ferrio et al., 2005; Riehl et al., 2014; Wallace et al., 2013, 2015). Studies have reported $\Delta^{13}C$ values of barley ranging between 16.2 to 18.0‰ during mid to late Holocene (~ 3000–1800 BCE) (Aguilera et al., 2008, 2012; Ferrio et al., 2005; Flohr et al., 2011; Mora-

Gonzalez et al., 2016; Reihl et al., 2014; Wallace et al., 2013, 2015). Araus et al. (1997) suggested $\Delta^{13}C$ value ~ 18‰ for barley crop imply irrigation practices in the arable fields (Mora-Gonzalez et al., 2016). However, $\Delta^{13}C$ values lesser than 16–17‰ are considered as a moderately poor water availability. Average $\Delta^{13}C$ values for barley grains grown during the early phase were found to be 15.3‰±2.0, indicating marginally drier conditions compared to those during transitional phase (~ 16.9‰±1.7) and the mature phase (~ 16.7‰±1.8). Hence, it appears to be an entirely different story for hydrological status of the Indus agricultural produce with respect to growing aridity in the region (Dixit et al., 2014; Enzel et al., 1999; Giosan et al., 2012; Prasad et al., 2014; Sarkar et al., 2016; Singh et al., 1974). $\Delta^{13}C$ values of cereals grown during the mature phase were found to be higher, thus indicate that the farming strategies has been practiced in an arid Indus region probably anthropogenically managed (rain fed + irrigation) (Wallace et al., 2013) due to the increasing dryness in temporal domain (during end part of the mature phase). The study of the oxygen isotope of two foraminifer species *Neogloboquadrina dutertrei* and *Globigerinoides sacculifer* from the Indus River delta in the Arabian Sea also provided the evidence for the seasonal changes in the Indus region (Giesche et al., 2019). This study recorded strong winter monsoon between 4.5 to 4.3 kyr and reduction in both winter and summer rainfall during 4.1 Kyr and also suggest the growth of Indus urban centres coincided with increased winter rainfall, however, the de-urbanism and change in subsistence strategies followed a reduction in both winter and summer season rainfall (Giesche et al., 2019). Thus, the $\Delta^{13}C$ values of barley grown during the mature phase in growing aridity supports our hypothesis of farming strategies such as water management or river channelling followed by the Indus farmers (for site chronology please see supplementary Table 1S). While Giosan et al., (2018) provided evidence for the stronger winter monsoon ~ 4.5 and 3.0 Kyr and Kathayat et al. (2017) provided evidence for the drier conditions between ca. 4.3 and 3.3 Kyr from the detailed reconstruction of summer monsoon in the Harappan domain. Our macro-botanical data (Sharma et al. 2020a) clearly indicated that Harappan farmers shifted their cropping strategy from winter crops to summer crops by growing millets, cotton and leguminous crops like *Vigna* sp.

Average $\Delta^{13}C_{cotton}$ values varied in the range 17.9‰±1.9 for the transitional phase and 16.5‰±2.6 for the mature phase (Table 1), indicating well-watered conditions (rain fed via summer precipitation + irrigation) during the transitional phase. It may be due to change in course of summer monsoon that reached the western Rajasthan Indus farmlands (~ 2600 – 2500 BCE) and supported crop-diversification. Summer monsoon however, appears to have subsequently declined toward late mature phase as evidenced by arrival of millet based cropping pattern (Sharma et al., 2020a). Similar observations have also been made in our earlier studies (Pokharia et al., 2017). We surmise that the Harappan farmers exploited both the seasonal rainfall and other means of water management in arable lands such as steering river waters in their agricultural fields. More isotopic data of archaeological grains from other peripheral archaeological sites are needed to garner much deeper insights into the Indus farming especially for the latter part of the mature phase (~ 1900–1800 BCE) of the Indus era.

$\delta^{15}N$ values of the crop-remains in conjunction with soil-sediment have been used to assess aridity in farmlands and agricultural intensifying efforts (*e.g.* manuring) (Agnihotri et al., 2021; Bogaard et al., 2007; Kanstrup et al., 2012; Styring et al., 2017). $\delta^{15}N$ values of host soil-sediment largely reflect land-use history; larger the magnitude greater the reworking of arable soils (Peukert et al., 2012). Enhanced $\delta^{15}N$ values of host soil-sediment could also be due to enhanced aridity (loss of water taking out lighter isotope bound nitrogen from soil). Manuring of farmlands with animal dung generally result in enhanced $\delta^{15}N$ values of host soil-sediments (Bol et al., 2005; Szpak et al., 2014). For instance, the effect of cattle dung manure in farmlands recorded $\delta^{15}N$ values ranging between + 2‰ to + 8‰, while the values for pig manure range between + 15‰ to + 20‰ (Bateman and Kelly, 2007; Szpak et al., 2014).

To deduce the factual interpretation of $\delta^{15}N$ data, therefore, Nitrogen concentration data (TN %) greatly help deciphering various aforementioned processes. Aridity conditions would favour higher $\delta^{15}N$ values with lower TN contents, while organic manuring would result in higher TN contents (due to richness of nitrates and ammonia in soils) along with higher $\delta^{15}N$ values (Fuertes-Mendizábal et al., 2018; Gron et al., 2021; Szpak, 2014). TN contents in farmlands could also enhanced by cropping leguminous plants (*e.g.* pulses) that fix nitrogen from atmosphere. In this kind of agricultural intensification, agricultural grains/seeds would show enhanced TN contents but lower $\delta^{15}N$ values (as $\delta^{15}N$ of atmospheric N_2 is regarded as 0.0‰; van Klinken et al., 2000). Similar effect would be seen in case of application ammonia based fertilizers (urea) as these contain large pool of reactive Nitrogen that has been fixed from atmospheric source (using Haber process) (Bateman et al., 2005). This case, however, is applicable only in modern day agricultural fields as this practice was introduced in India after 1970's.

Archaeological barley grains from the studied site (4MSR) show an enhancing trend in $\delta^{15}N$ values in terms of its intra-phase variability (as determined by standard deviation) from early to mature phase. Macro-botanical data revealed a conspicuous crop-diversification was conducted during the short transitional phase via adopting leguminous crops (pulses) (Sharma et al., 2020a). This short duration of transitional phase also witnessed marginally enhanced TN contents ($p = 0.084$) (Table 1). This observation could be interpreted in terms of successful crop-shifting strategy adopted by Harappan farmers to enhance soil nutritional health and their agricultural produce during the transitional phase which continued towards the end of mature phase. As a matter of fact, a significant diversification of crops (towards pulses) was inferred with two fold enhancement (in abundance) from the early to transitional phase, and further two fold enhancement from transitional to mature phase (using macro-botanical data from this site earlier; Sharma et al., 2020a).

Earlier in controlled agronomic conditions, Bogaard et al. (2013) quantified ancient manuring practice from the measured $\delta^{15}N$ values of modern bulk cereal samples (barley and wheat) cultivated in Europe and noted higher $\delta^{15}N$ values (ranging from 6–9‰) correspond to high manuring rates (≥ 35 + tons/hectare) while lower values (≤ 3.0‰) corresponds to long-term unmanured cultivation. Fraser et al., (2011) also recoded $\delta^{15}N$ values of modern bulk cereal samples (wheat and barley) grown in Germany under different manuring rates and quantified $\delta^{15}N$ values (1) 0.0 to 3.0‰ as low manuring, (2) ~ 3.0 to 6.0‰ as medium manuring and (3) above 6.0‰ as high manuring. Nitsch et al. (2017) published $\delta^{15}N$ values of barley ranging between 5.4 to 6.8‰ for Early Bronze Age and 4.0‰±1.2 for Late Bronze Age from Archontiko. In realm of all above mentioned datasets, we place our crop isotopes dataset that supports possibly a combined manifestation of crop-management through diversification, for instance, by cultivating more leguminous crops.

Conclusions

Following conclusions could be drawn from the study-

- The $\delta^{13}C$ values of both winter and summer crops (barley and cotton) do not indicate any significant difference in the hydrological conditions among different Indus phases (early, transitional and mature phase) indicating Indus farming strategies to overcome deteriorating summer monsoonal conditions during increased dryness in the latter part of the mature phase.
- $\delta^{13}C$ values of soil organic matter however clearly indicate a progressive regional aridity *i.e.* a transition from wetter to drier conditions. The growing aridity was found to have well corroborated by the crop-assemblages data that recorded dominance of millet based crops during the latter part of mature phase (Sharma et al., 2020a).
- Observed scenario indicates that the Harappan farmers were likely managing working hydrological status by other means such as channelling river waters into their agricultural fields.
- $\delta^{15}N$ soil-sediments (along with Total Nitrogen contents) indicate progressive agricultural intensification from early to mature phase, which yielded better crop-quality and soil-fertility under prevailing aridity.
- Lower $\delta^{15}N$ values (with enhanced Total Nitrogen contents) of weed (tiny vetch) demonstrate plausible impact of crop-diversification and efficacy of crop-management by sowing pulses during the mature phase.
- It appears that Harappan farmers were capable of changing their farming strategies during the mature phase.

Declarations

Author Contributions

Conceptualization: SS, RA. Data curation: SS, RA. Investigation (data collection and analysis): SS, RA, AKP, AK. Writing–original draft: SS, RA. Writing — review & editing: SS, RA, AKP, AK, RB. SKM performed excavation.

Competing interest

The authors declare no competing interest.

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Data availability

All data are availability in the main text and the supplementary information of the manuscript.

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Figures

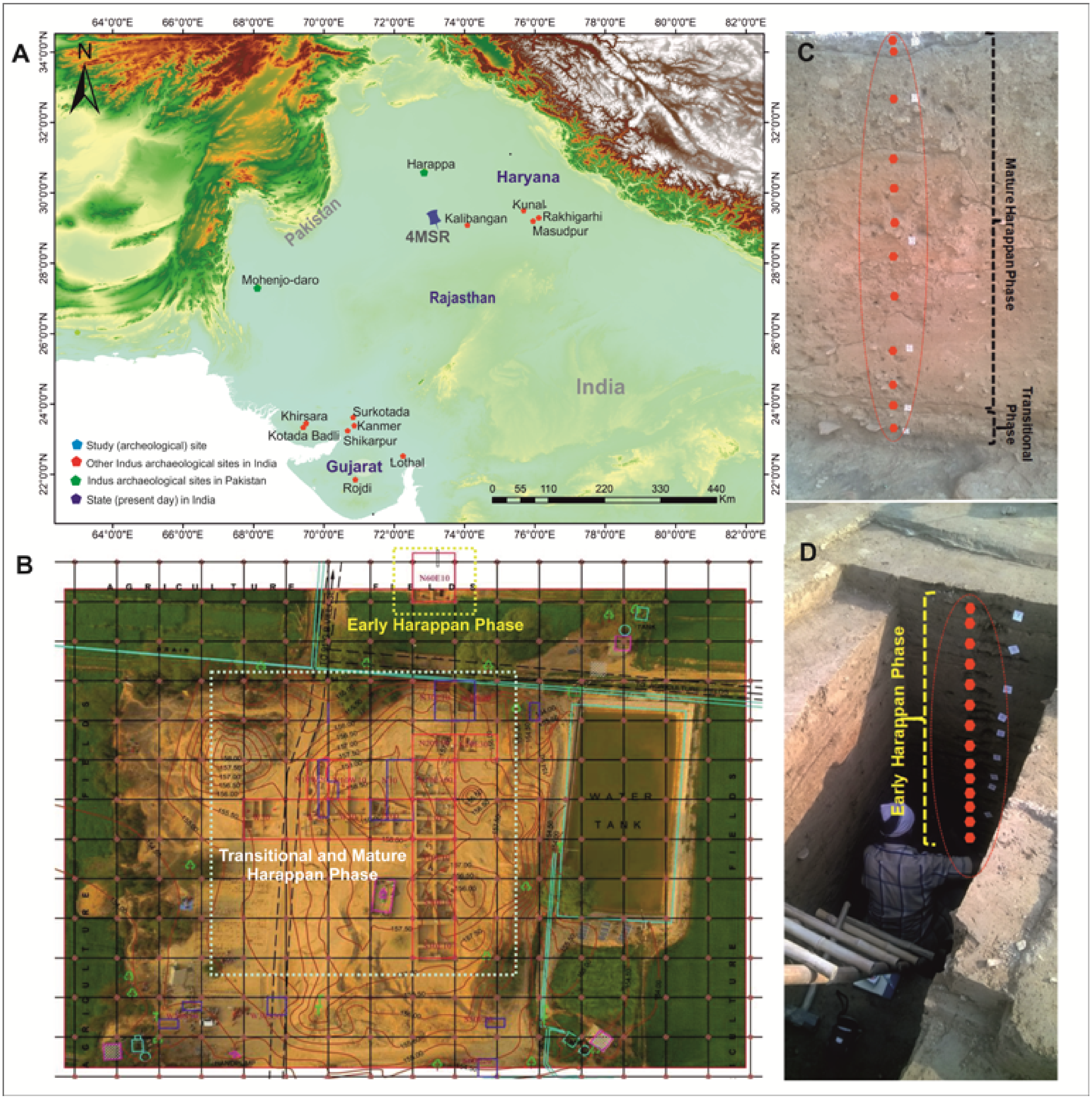

Figure 1

(A) Map showing location of archaeological study site 4MSR, Rajasthan along with other Indus sites (created by using mapping software *ArcGIS 10.3*). (B) Contour map of the study site shows excavated trenches and upper right shows the aerial view of the excavated site taken by dronne. (C) Section shows the location of sample collection from the mature Harappan and transitional phase stratigraphic layers. (D) Trench representing the sample collection area from the early Harappan phase deposit.

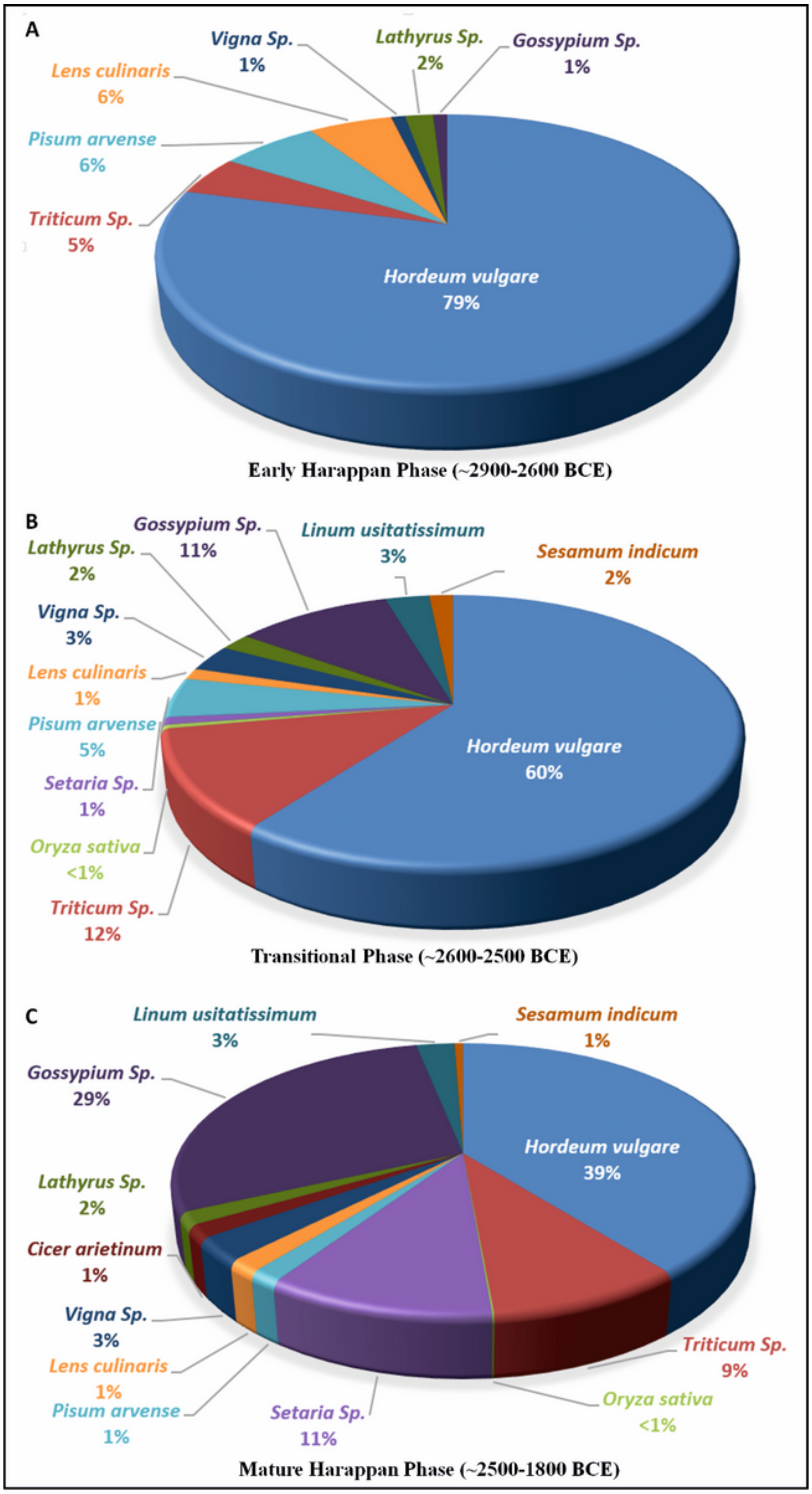

Figure 2

Pie charts showing the relative proportion of crop species during (A) early phase, (B) transitional phase, and (C) mature phase of Indus era recovered from the site 4MSR (modified from Sharma et al., 2020a).

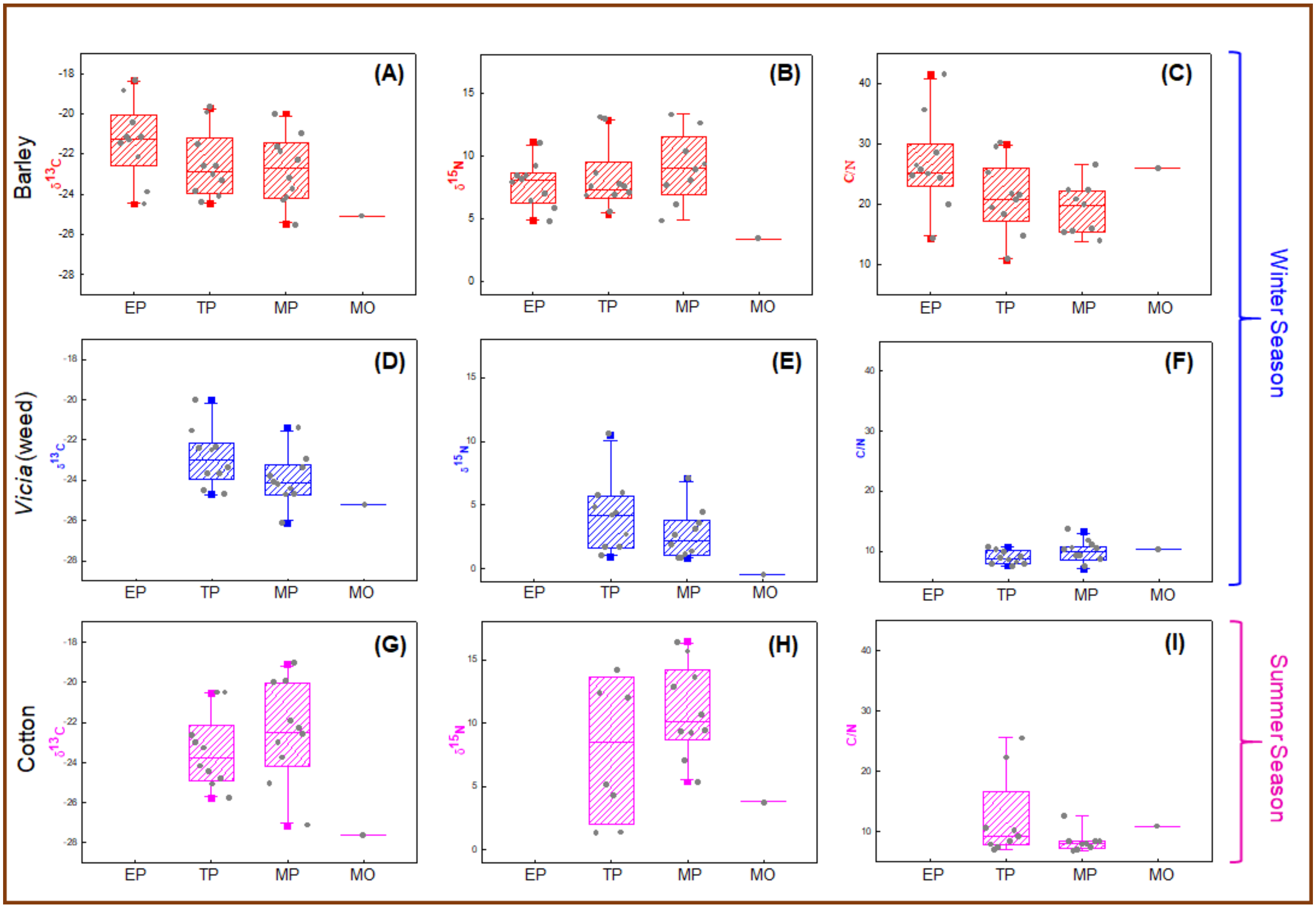

Figure 3

Box-Whisker plots showing the d[13]C (A, D, G), d[15]N values (B, E, H), and C/N ratios (C, F, I) of barley, *Vicia* (common/tiny vetch) and cotton respectively, recovered from different cultural phases from the archaeological site 4MSR. Data points (shown in grey dots) display number of seeds/grains measured (*n*=10; for each crops and weed). The line inside the box represents median, upper half of the box represents upper quartile, while the lower half represents lower quartile. (Abbreviations: EP= Early Harappan Phase; TP= Transitional Phase; MP= Mature Harappan Phase; MO= Modern).

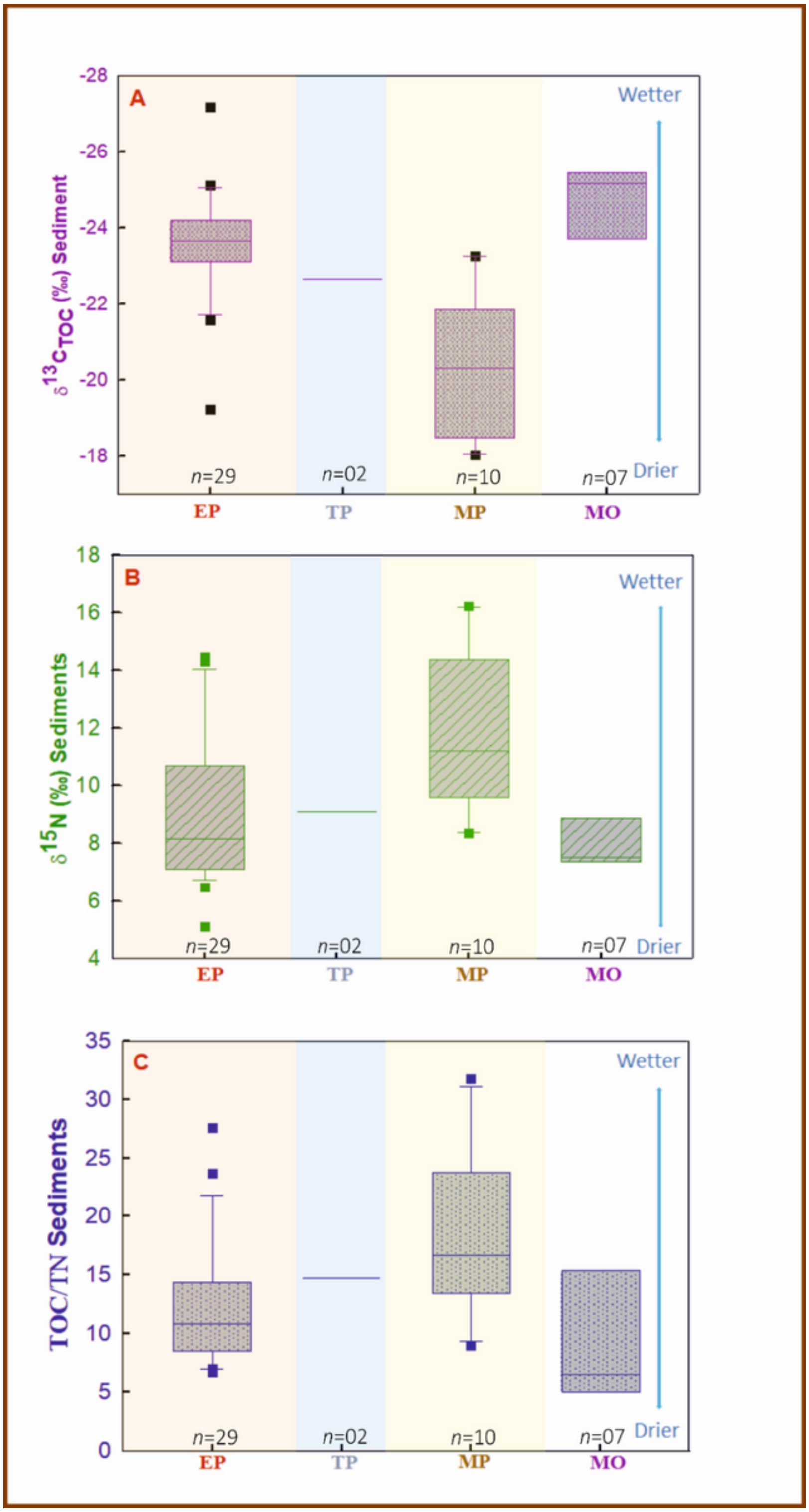

Figure 4

Box-Whisker plots showing (A) d^{13}C$_{SOM}$, (B) d^{15}N$_{Sediment}$ values, and (C) C/N$_{Sediment}$ ratios of soil-sediments from different cultural phases. The line inside the box represents median, upper half of the box represents upper quartile, while the lower half represents lower quartile. The values lies outside the min and max range are outliers. (Abbreviations: EP= Early Harappan Phase; TP= Transitional Phase; MP= Mature Harappan Phase; MO= Modern).

Supplementary Files

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